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THEORY OF PRESENT AND FUTURE SAFEGUARDS
APPLICATIONS OF ISOTOPIC RATIOS

CONTRACT NUMBER ACDA/WEC-216

PREPARED FOR
U.S. ARMS CONTROL AND DISARMAMENT AGENCY

PREPARED BY

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by

D. L. Prezbindowski

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THEORY OF PRESENT AND FUTURE SAFEGUARDS APPLICATIONS OF ISOTOPIC RATIOS

D. L. Prezbindowski

Abstract

Previous studies⁽¹⁾ have identified certain isotopic ratios which are promising for use in verifying the Pu content in the nuclear fuels dissolved at the chemical reprocessing plant. This is especially true with ratios which are highly constant over a large range of fuel exposure. The usefulness of these isotopic ratios is that they provide a basis for inter-comparing isotopic data of a wide range of exposure.

One of the contributions that theory has made in the development of isotopic ratios is to reveal the physical basis for the invariance of the ratio $\text{Pu/U}/^{235}\text{D}$ in U enriched PWR fuel through simplified transmutation models. Although the simplified models give insight into the reasons for the constancy of $\text{Pu/U}/^{235}\text{D}$ and a qualitative agreement with experimental data, they are not accurate and general enough to be used as a quantitative tool for safeguards.

Before an accurate and general approach to the problem of constructing isotopic ratios can be defined the important question of the sensitivity of an isotopic ratio to diversion must be examined. Any one isotopic ratio will have zero sensitivity to certain types of diversion; this zero sensitivity is called a "blind spot." Methods of identifying and compensating for "blind spots" in the isotopic ratio require the solution of a system of equations which predict the possible combinations of small isotope changes

that will produce no response in the isotopic ratios being used. The use of a number of distinct ratios that is greater than the number of isotopic concentrations that the diverter may conceivably manipulate insures that the "blind spots" to diversion are covered. Two or three ratios may be all that are required in practice since other constraints may eliminate many mathematically possible but practically impossible diversions.

The process of constructing isotopic ratios has two objectives:

- The definition of the ratio
- The specification of the characteristics of the ratio over a variation of reactor parameters that influence the production and depletion of isotopes being safeguarded.

The construction process achieves these goals using the most accurate measured and calculated data available. The present accuracy of calculational methods is sufficient to permit them to play an important part in the development of isotopic ratios for future applications. Even the most careful construction of isotopic ratios generally leaves undesirable limitations on the constancy and range of application of the ratios.

A whole new dimension for improving the constancy and reliability of the isotopic ratio method is the use of isotopic powers and coefficients to improve the isotopic ratio. The use of isotopic powers and coefficients is equivalent to having a limited capacity to adjust the basic nuclear cross sections. Examples of the application of coefficients show that a ratio ($\text{Pu}/\text{U}/^{235}\text{D}$) with application to U^{235} enriched PWR cores with small natural background variations (4 percent or less) can have these variations reduced by a factor of 1000. A ratio with applications to BWR's with larger compound variations (34 percent or less) has had these variations reduced to one percent or less over specified ranges of exposure and moderator void.

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THEORY OF PRESENT AND FUTURE SAFEGUARDS APPLICATIONS OF ISOTOPIC RATIOS

D. L. Prezbindowski

I. INTRODUCTION

The decision to use isotopic ratios must be based on a good understanding of their behavior as it is related to the safeguards function. It is especially important to understand how to define isotopic ratios which have the highest degree of constancy over prescribed ranges of reactor perturbations and exposure. The reliance to be placed on isotopic ratios must be assessed with the help of theory by determining whether there is a methodology based on theory that will produce isotopic ratios with the desired characteristics in future applications. The purpose of this paper is to present some insights into the constancy of certain isotopic ratios with potential safeguards applications, and also to present some methods of making isotopic ratios more accurate and foolproof in safeguards applications.

The first section of the paper presents discussions which are designed to give some insight into isotopic ratios, especially the isotopic ratio $Pu/U/^{235}D$. The approach used is generally one of simplifying the burnup calculational model in order to reveal the underlying reasons for the constancy of these ratios with exposure in PWR's. The approximations made in the simplification process introduce errors which limit the application of the simplified model. Two approaches to understanding the constancy of the ratio as applied to the YANKEE ROWE PWR are presented which represent two different balances between accuracy and simplicity.

The second section of the paper presents methods of constructing, improving, and establishing the reliability of isotopic ratios that are quantitatively accurate. This methodology is based on the theory describing isotopic ratios.

The Appendix contains mathematical details from Section I.

II. SUMMARY AND CONCLUSIONS

The ideas that are presented in this paper systematically outline the means of dealing with present and future applications of isotopic correlation safeguard techniques through theoretical methods. They also provide a rational basis for constructing isotopic ratios useful to such techniques and for identifying their ranges of applicability. This work has been undertaken so that the strengths and limitations of the isotopic ratio methods for safeguards application will be well understood.

Previous studies^(1,2,3,4) have identified certain isotopic ratios that are promising for use in verifying the Pu content of nuclear fuels as measured at a chemical reprocessing plant. A characteristic of the ratios is that they are highly constant over a large range of fuel exposure. The usefulness of these isotopic ratios is that they provide a basis for intercomparing isotopic data of a wide range of exposure. The ability to verify the internal consistency of the data and to confirm agreement with measured standards via isotopic ratios substantially reduce the number of independent measurements needed for safeguards.

One of the contributions that theory has made is to provide insight and understanding into the invariance of the isotopic ratio $\text{Pu}/\text{U}/^{235}\text{D}$ in U enriched light water reactors through simplification of the isotopic transmutation model. For purposes of simplification of the theoretical analysis, the ratio $\text{Pu}/\text{U}/^{235}\text{D}$ (where ^{235}D is the ^{235}U depletion in weight percent,) has been related to the simplified ratio $\text{Pu}/^{235}\text{d}$, (where ^{235}d is the ^{235}U depletion in terms of number of atoms). The analysis has been performed on this simplified ratio.

Theory predicts that the slope of the Pu versus ^{235}d curve can be expressed approximately in the form $C [a - x/b - x]$ where C is a constant, a is the ^{238}U neutron capture rate, x is the sum of the ^{239}Pu and ^{241}Pu fission rates, and b is the initial ^{235}U energy production rate times the ratio of the initial and final neutron flux levels. At the zero irradiation point, in a U enriched reactor, the Pu concentration

is zero and the slope expression reduces to the initial conversion ratio. Expressions for the sensitivity of the slope to variations in a , b and x have also been derived.

The knowledge of the limits of the linearity of $Pu/U/^{235}D$ is necessary for the successful application of this ratio for safeguards purposes. The variation in $Pu/U/^{235}D$ is reduced to about 0.5% in the exposure range 7,500-25,000 MWd/T for YANKEE ROWE data if ^{242}Pu is not included in the definition of $Pu/U/^{235}D$. If ^{242}Pu is included in the definition the variation is about 3% over the range 7,500-25,000 MWd/T.

The derived expression for the slope is in agreement with experimental and calculated results with regard to the influence of ^{235}U enrichment and neutron flux energy spectrum. The slope expression is therefore a useful formula for estimating the influence on the slope of enrichment and flux spectrum changes due to changes in moderator to fuel ratio, cladding and fuel dimensions.

The usefulness of the derivation also lies in the insight obtained into the physical reason for the observed linearity of the ratio $Pu/U/^{235}D$ for various reactor cores. Now that one can determine the degree of linearity to be expected, and the parameters affecting the slope have been identified, it is much easier to extrapolate the isotopic correlation method to reactor cores where no historical data exists. In addition, one can estimate how various reactor design and operation variables will influence the slope.

Although the simplified models give insight into the reasons for the constancy of $Pu/U/^{235}D$ and give a qualitative agreement with experimental data, the models are not accurate and general enough to be used as a quantitative tool for safeguards.

Before an accurate and general approach to the problem of constructing isotopic ratios can be defined, the important question of the sensitivity of an isotopic ratio to diversion must be examined. Any one isotopic ratio will have zero sensitivity to certain types of diversion; this zero sensitivity is called a "blind spot." Methods of identifying and compensating for "blind spots" in the isotopic ratio require the solution of a system of equations which predict the possible combinations

of small isotope changes that will produce no response in the isotopic ratios being used. The use of a number of distinct ratios that is greater than the number of isotopic concentrations that the diverter may conceivably manipulate insures that the "blind spots" to diversion are covered. Two or three ratios may be all that are required in practice since other constraints may eliminate many mathematically possible but practically impossible diversions.

The process of constructing isotopic ratios has two objectives:

- The definition of the ratio
- The specification of the characteristics of the ratio over a variation of reactor parameters that influence the production or depletion of isotopes being safeguarded.

The construction process achieves these goals using the most accurate measured and calculated data available. The present accuracy of calculational methods is sufficient to permit them to play an important part in the development of isotopic ratios for future applications.

Even the most careful construction of isotopic ratios generally leaves undesirable limitations on the constancy and range of application of the ratios. A whole new dimension for improving the constancy and reliability of the isotopic ratio method is the use of isotopic powers and coefficients to improve the isotopic ratio. The use of isotopic powers and coefficients is equivalent to having a limited power to adjust the basic nuclear cross sections. Examples are given of the application of coefficients in the ratio $(\text{Pu}/\text{U}/^{235}\text{D})$ for ^{235}U enriched PWR cores and in the ratio $[\text{Pu}/(^{235}\text{U} + ^{236}\text{U})]$ Pu enriched PWR cores. The small natural background variations (4% or less) can be reduced by a factor of 1000. A ratio with applications to BWR's with larger compound variations (34% or less) has been developed to reduce these variations to 1% or less over specified ranges of exposure and moderator void.

III. INVARIANCE OF Pu/U/²³⁵D WITH EXPOSURE

GENERAL APPROACH

Previous studies^(1,2,3,4) have identified certain ratios which are promising for use in safeguards applications, especially ratios which result from relationships which are highly linear over a large range of exposure. The ratio, Pu/U/²³⁵D (²³⁵D is the change in ²³⁵U enrichment during irradiation or ²³⁵U depletion), has shown a high degree of linearity from measured data (see Figures 1 and 2). The linear relationship mentioned has been observed in U and Pu total concentrations and isotopic data where the data resulted from measurements made on dissolved spent fuel at the accountability tank of a chemical reprocessing plant. The data and the relationships evidenced by the data represent large portions of a reactor core, as large as a metric tonne of spent fuel per batch of dissolved spent fuel (that is, from 2 to 16 fuel assemblies, depending on size, were dissolved at a given time). The relationships are thus averages of the various conditions that actually existed at different locations in a fuel assembly during irradiation.

The isotopic ratio of Pu/U versus ²³⁵D is one which is very promising from the point of view of safeguards applications. As shown in Figures 1 and 2 the experimental plots of this ratio of YANKEE ROWE,^(2,4) DRESDEN,⁽²⁾ and CANDU⁽⁸⁾ cores are highly linear. These data are obtained from the dissolution of fuel from several cores of the reactors concerned. It is useful to analyze the reasons for this linearity so that the reliability of the phenomena can be determined and the sensitivity of the slope of the linear function to various factors can be determined. The definitions of Pu/U and ²³⁵D as plotted in the figures have been chosen for convenience of application to safeguards. Specifically, ²³⁵D is defined as follows:

FIGURE 1. Pu/U Ratio Versus ^{235}D Resulting from Chemical Reprocessing Plant Measurements. (The curves were drawn in by inspection.)

FIGURE 2. Pu/U Ratio Versus ^{235}U Weight Percent. (This presentation of the data given in Figure 1 conveniently separates the plotted points by initial enrichment. The curves were drawn in by inspection.)

$$^{235}\text{D} = \left(\frac{\text{grams } ^{235}\text{U}}{\text{grams } ^{238}\text{U} + \text{grams } ^{235}\text{U} + \text{grams } ^{236}\text{U} + \text{grams } ^{234}\text{U}} \right) \text{ before irradiation} \\ - \left(\frac{\text{grams } ^{235}\text{U}}{\text{grams } ^{238}\text{U} + \text{grams } ^{235}\text{U} + \text{grams } ^{236}\text{U} + \text{grams } ^{234}\text{U}} \right) \text{ at time } t \quad (1)$$

and

$$\text{Pu/U} = \left[\frac{\text{total Pu (grams)}}{\text{total U (tonnes)}} \right] \text{ at time } t \quad (2)$$

Another way of expressing ^{235}D is:

$$^{235}\text{D} = \text{initial enrichment (in weight percent)} \\ - \text{final enrichment (in weight percent)}.$$

The initial enrichment can be determined, as defined above, from the fuel fabricator's accountability sheets. The definitions of final enrichment and Pu/U are consistent with the data available from the chemical reprocessor.

These definitions can be simplified for the purposes of investigating linearity. Definitions which are simpler and more usable from a calculational viewpoint are as follows:

$$^{235}\text{d} = (N^{235})_{\text{before irradiation}} - (N^{235})_{\text{at time } t} \quad (3)$$

$$\text{Pu} = (N^{239} + N^{240} + N^{241} + N^{242})_{\text{at time } t} \quad (4)$$

where

$$N^g = \text{number of atoms of isotope } g \text{ per cubic centimeter} \quad (5)$$

A small "d" is used in definition (3) to differentiate between that definition and definition (1). Definition (1) can be rewritten in a way which reveals the relationship between definition (1) and definition (3):

$$^{235}\text{D} = \frac{G^{235}}{\text{grams/cm}^3 (^{238}\text{U} + ^{235}\text{U} + ^{236}\text{U} + ^{234}\text{U}) \text{ before irradiation}}$$

$$\left[(N^{235}) \text{ before irradiation} - K(N^{235}) \text{ at time } t \right] \quad (6)$$

where

G^{235} = grams of ^{235}U per atom of ^{235}U

$$K = \frac{\text{grams/cm}^3 (^{238}\text{U} + ^{235}\text{U} + ^{236}\text{U} + ^{234}\text{U}) \text{ before irradiation}}{\text{grams/cm}^3 (^{238}\text{U} + ^{235}\text{U} + ^{236}\text{U} + ^{234}\text{U}) \text{ at time } t} \quad (7)$$

The factor K initially has the value 1.0 and then it increases about 4% over the exposure range 0-25,000 MWd/MTU in YANKEE ROWE. (That is to say 4% of the initial total U disappears during irradiation.) The ratio in front of the brackets in Equation (6) is a constant with exposure and cannot affect the linearity of $\text{Pu/U}/^{235}\text{D}$ as a function of exposure.

An important step in establishing the reliability and the range of invariance of the ratio $\text{Pu/U}/^{235}\text{D}$ is to analyze and identify the physical basis for the observed linearity. A necessary and valid simplification of the analytical approach of studying the relationship from a batch of discharged reactor fuel is to study the relationship at a specified point in a fuel bundle. If linear relationships are established for each point in the fuel bundle then the superposition of the linear behavior at all points in the fuel results in a linear relationship for the average.

The derivation will be supported by calculated data from YANKEE ROWE Cores I through VI. The calculation of the depletion of the reactor fuel in YANKEE ROWE was performed using the computer code ZODIAC-G.⁽⁹⁾ In addition, experimental data from YANKEE ROWE Cores I through VII and several other reactor cores have been presented which demonstrate the empirical basis for the observed linearity. The intent is to show that

the linearity can be supported via theory on a point basis and that the linearity does indeed occur when one looks at data averaged over an entire core.

As a first step in deriving an expression for $Pu/^{235}d$, some previous work in this regard will be reviewed. Then a new derivation will be presented which is felt to have greater applicability to power reactor cores. The new derivation will first develop an expression for the slope of the line relating Pu to ^{235}d , and then discuss how this relates to $Pu/U/^{235}D$ which is the ratio used in MIST.

B. PREVIOUS DERIVATION

C. R. Weisbin and M. M. Thorpe have documented a theoretical discussion of the linearity of Pu versus ^{235}d .⁽¹⁰⁾ They have expanded the number density of ^{239}Pu as a function of ^{235}U depletion as follows:

$$\begin{aligned} \frac{N^{239}(t)}{N^{238}(o)} = & \frac{\frac{-238}{\sigma_c}(t)}{\frac{-235}{\sigma_a}(t)} \left[\frac{^{235}d}{N^{235}(o)} - \left(\frac{\frac{-239}{\sigma_a}(t) - \frac{-235}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{^{235}d}{N^{235}(o)} \right)^2 \right. \\ & + \left(\frac{\frac{-239}{\sigma_a}(t) - \frac{-235}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{\frac{-239}{\sigma_a}(t) - 2\frac{-235}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{^{235}d}{N^{235}(o)} \right)^3 + \dots \left. \right] \\ & + \frac{N^{239}(o)}{N^{238}(o)} \left[1 - \left(\frac{\frac{-239}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{^{235}d}{N^{235}(o)} \right) \right. \\ & + \left(\frac{\frac{-239}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{\frac{-239}{\sigma_a}(t) - \frac{-235}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{^{235}d}{N^{235}(o)} \right)^2 \\ & - \frac{\frac{-239}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \left(\frac{\frac{-239}{\sigma_a}(t) - \frac{-235}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{\frac{-239}{\sigma_a}(t) - 2\frac{-235}{\sigma_a}(t)}{\frac{-235}{\sigma_a}(t)} \right) \left(\frac{^{235}d}{N^{235}(o)} \right)^3 + \dots \left. \right] \quad (8) \end{aligned}$$

A number of the assumptions used in their analysis are as follows:

- (1) There are only two fissionable isotopes, ^{235}U and ^{239}Pu in the reactor.
- (2) The burnup of ^{238}U is negligible.
- (3) The group averaged cross sections are slowly varying functions of time, particularly the effective absorption cross-section of ^{235}U and the effective capture cross section of ^{238}U .

Their argument for linearity of $\frac{N^{239}(t)}{N^{238}(o)}$ versus ^{235}d is based on the fact that if Equation (8) is divided through by ^{235}d and $N^{239}(o) = 0$ (as in the case in U enriched LWR's), then

$$\frac{N^{239}(t)}{N^{238}(o)} / ^{235}\text{d} \approx \left(\frac{\bar{\sigma}_c^{238}(t)}{\bar{\sigma}_a^{235}(t)} \right) \left(\frac{1}{N^{235}(o)} \right) \quad (9)$$

for ^{235}U depletion small enough to permit the neglect of the second and higher terms in the expansion. Under the assumption that $\bar{\sigma}_c^{238}(t)$ and $\bar{\sigma}_a^{235}(t)$ are constant, then

$$\frac{N^{239}(t)}{N^{238}(o)} / ^{235}\text{d}$$

is approximately constant. The assumption of a constant $\bar{\sigma}_a^{235}(t)$ and a small ^{235}d limits the applicability of this analysis for certain reactors of interest. Figure 3 shows the variation of $\bar{\sigma}_a^{235}$ as a function of exposure in the asymptotic zone of YANKEE ROWE Cores I and II as calculated by the ZODIAC-G program. The percent variation of this cross section is 23% over the exposure range of 25,000 MWd/MTU. At 25,000 MWd/MTU

the $^{235}\text{d}/N^{235}(0)$ term is about 0.6 in YANKEE ROWE so that the second term in the expansion of Equation (8) is about 30% of the first and cannot be entirely neglected.

FIGURE 3. σ_a^{235} Versus Exposure Calculated by ZODIAC for Yankee Rowe

C. NEW DERIVATION

In order to overcome some of these problems an effort was undertaken to develop a theoretical basis for linearity of Pu versus ^{235}d which has broad applicability with regard to reactor type and considers all of the four major plutonium isotopes, ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu . Furthermore, no restrictive assumptions will be made with regard to the burnup of any isotope or with regard to the variation of any effective cross sections as a function of exposure. Pu radioactive decay processes are assumed to be negligible.

The detailed derivation is given in Appendix A. The general approach used was to derive a differential equation expressing the rate of change of the total plutonium concentration with respect to ^{235}d . The observation is made that in the hypothetical case where ^{235}U is the only fissionable isotope present there will be a highly linear relationship between ^{235}D and exposure as shown in Figure 4. However, the buildup of Pu in a uranium enriched reactor causes a significant non-linearity in the ^{235}D as a function of exposure as shown in Figure 4. As the Pu builds up it also fissions and produces power so that at any particular value of total power generated, (MWd), the depletion of ^{235}U is less than it would have been in the absence of Pu. In this way, the Pu buildup exerts its influence on the ^{235}U depletion. This relationship is expressed in a conservation of power expression which is manipulated and combined with the differential equation expressing the rate of change of total Pu as a function ^{235}U depletion. This combination produces a slope expression in the form $C [a - x/b - x]$ where C is a constant, a is the ^{238}U capture rate, x is the sum of the ^{239}Pu and ^{241}Pu fission rates, and b is the initial ^{235}U energy production rate times the ratio of the initial and final flux levels. Plots of a, b, and x are given in Figures 5 and 6.

At the zero irradiation point in a ^{235}U enriched reactor the Pu concentration is zero and the slope expression reduces to the initial conversion ratio. Expressions for the sensitivity of the slope to variations in a, b, and x are also derived. A comparison of the variations in the slope of Pu versus ^{235}d and Pu/U versus ^{235}D in the exposure

FIGURE 4. ^{235}D and Pu/U Versus Exposure

FIGURE 5. Calculated $x(t)$ Versus Exposure for Yankee Rowe Cores I and II

FIGURE 6. $N_{(o)}^{235} \sigma_{f(o)}^{235} \frac{\phi(o)}{\phi(t)}$ and $N_{(t)}^{238} \bar{\sigma}_c^{238}$ Versus Exposure

range 7,500-25,000 MWd/MTU shows that the choice of $\text{Pu/U}/^{235}\text{D}$ from practical safeguards considerations has been a judicious choice resulting in a cancellation of much of the variation found in the ratio $\text{Pu}/^{235}\text{d}$.

The knowledge of the limits of the linearity of $\text{Pu/U}/^{235}\text{D}$ establishes the degree of accuracy that one may expect from this safeguards tool. For example, at high exposures, >30,000 MWd/MTU, one should expect a slight increase in the slope of the line. With the slope increase identified, it is easy to account for it. Also, the initial ^{235}U enrichment and flux spectrum have a significant influence on the linearity property. The initial ^{235}U enrichment in D_2O and H_2O moderated reactors range from the 0.7% enrichment found in natural U to between 4 and 5% in enriched U in H_2O moderated reactors. The variation of the initial ^{235}U enrichment will affect b. $\text{Pu/U}/^{235}\text{D}$ will vary inversely with the value of the initial ^{235}U enrichment. There will be some compensating variation in the microscopic fission cross section of ^{235}U due to flux spectrum changes. This influence of initial enrichment on the slope of the $\text{Pu/U}/^{235}\text{D}$ is shown experimentally in Figure 7. It can be seen from Figure 7 that the various reactors are ranked according to enrichment by the slope. The one exception is the VAK⁽⁸⁾BWR where it appears the effect of the flux spectrum is sufficient to change the ordering of the reactor by enrichment. The effect of decreasing the effective ^{235}U fission cross section through increasing the void fraction is shown in Figure 8. Increasing the void fraction hardens the flux spectrum through a decrease in moderator to fuel ratio and produces the increase in slope. In both cases the curves conform to the functional dependence predicted by the slope expression. Other parameters which affect the flux spectrum, such as fuel dimensions and type of cladding, will also affect the ratio. However, their affect can also be investigated through the slope expression.

FIGURE 7. Pu/U Ratio Versus ^{235}U Depletion for Various ^{235}U Enrichments

FIGURE 8. Pu/U Versus ^{235}U Depletion Calculated by ALTHAEA for Various Void Fractions in the Moderator

D. Coefficients in Isotopic Ratios Containing Plutonium

The particular functional form of the Pu isotopic concentrations as a function of time in uranium enriched reactors suggests a method of improving the constancy of isotopic ratios containing plutonium by the use of appropriate coefficients. It is useful to present some observations on the functional form of the plutonium isotopic concentrations as a function of time in uranium enriched reactors which initially contain no plutonium. The plutonium isotopic concentrations are an integral part of many isotopic ratios designed to safeguard plutonium.

We can write the ^{239}Pu isotopic concentration as a function of time in a power series:

$$N^{239}(t) = C_0 + C_1 t + C_2 t^2 + C_3 t^3 + C_4 t^4 + \dots \quad (10)$$

The initial condition:

$$N^{239}(0) = 0 \quad (11)$$

requires that

$$C_0 = 0. \quad (12)$$

Therefore

$$N^{239}(t) = C_1 t + C_2 t^2 + C_3 t^3 + C_4 t^4 + \dots \quad (13)$$

The differential equation for ^{240}Pu is

$$\frac{dN^{240}(t)}{dt} = -N^{240}(t)\bar{\sigma}_a^{240}(t)\phi(t) + N^{239}(t)\bar{\sigma}_c^{239}(t)\phi(t) \quad (14)$$

assuming that $\bar{\sigma}_a^{240}$, $\bar{\sigma}_c^{239}$ and ϕ are constant and integrating the above equation we have:

$$N^{240}(t) = -\bar{\sigma}_a^{240}\phi \int_0^{t_0} N^{240}(t) dt + \bar{\sigma}_c^{239}\phi \int_0^{t_0} N^{239}(t) dt. \quad (15)$$

Let

$$N^{240}(t) = D_0 + D_1 t + D_2 t^2 + D_3 t^3 + D_4 t^4 + \dots \quad (16)$$

Substituting (16) into (15) and performing the indicated integrations we obtain:

$$\begin{aligned} D_0 + D_1 t_0 + D_2 t_0^2 + D_3 t_0^3 + \dots = & -\bar{\sigma}_a^{240}\phi \left(D_0 t_0 + D_1 \frac{t_0^2}{2} + D_2 \frac{t_0^3}{3} + D_3 \frac{t_0^4}{4} \dots \right) \\ & + \bar{\sigma}_c^{239}\phi \left(C_1 \frac{t_0^2}{2} + C_2 \frac{t_0^3}{3} + C_3 \frac{t_0^4}{4} \right) \end{aligned} \quad (17)$$

The initial condition:

$$N^{240}(0) = 0 \quad (18)$$

Requires that

$$D_0 = 0 \quad (19)$$

Equating the coefficients of t_0 across equation (17) we have:

$$D_1 = -\bar{\sigma}_a^{240}\phi D_0. \quad (20)$$

From (20) we conclude

$$D_1 = 0. \quad (21)$$

The power series expansion for $N^{240}(t)$ is:

$$N^{240}(t) = D_2 t^2 + D_3 t^3 + D_4 t^4 + \dots \quad (22)$$

The differential equations for $N^{241}(t)$ and $N^{242}(t)$ are the same form as that of $N^{240}(t)$ (see Equations 69, 70, and 71). The same kind of argument as that used with $N^{240}(t)$ can be used to show that:

$$N^{241}(t) = E_3 t^3 + E_4 t^4 + E_5 t^5 + \dots \quad (23)$$

and

$$N^{242}(t) = F_4 t^4 + F_5 t^5 + F_6 t^6 + \dots \quad (24)$$

For t small enough so that each of the plutonium isotopes can be approximated by one term in the series expansion we may write:

$$N^{239}(t) + N^{240}(t) + N^{241}(t) + N^{242}(t) \approx C_1 t + D_2 t^2 + E_3 t^3 + F_4 t^4 \quad (25)$$

The isotopic ratio $\text{Pu}/\text{U}/^{235}\text{D}$ can be written as $\text{Pu} \left(\frac{1}{(\text{U})(^{235}\text{D})} \right)$

If we let:

$$\text{Pu}(t) = A(t) = a_1 t + a_2 t^2 + a_3 t^3 + a_4 t^4 + \dots \quad (26)$$

and

$$(\text{U}(t))(^{235}\text{D}(t)) = B(t) = b_1 t + b_2 t^2 + b_3 t^3 + b_4 t^4 + \dots$$

$$(\text{It is clear that } b_0 = 0 \text{ since } ^{235}\text{D} = 0 \text{ at } t = 0.) \quad (27)$$

We may then write:

$$P_{U/U}/^{235}D = \frac{a_1 t + a_2 t^2 + a_3 t^3 + a_4 t^4 + \dots}{b_1 t + b_2 t^2 + b_3 t^3 + b_4 t^4 + \dots} \quad (28)$$

The isotopic ratio will be constant as a function of time if:

$$a_i = b_i \quad i = 0, 1, 2, 3 \dots \quad (29)$$

If we multiply each isotopic concentration in (25) by a coefficient and substitute it into (28) we have:

$$\begin{aligned} & \left(V N^{239}(t) + X N^{240}(t) + Y N^{241}(t) + Z N^{242}(t) \right) / (U) (^{235}D) \\ & \approx \frac{V C_1 t + X D_2 t^2 + Y E_3 t^3 + Z F_4 t^4}{b_1 t + b_2 t^2 + b_3 t^3 + b_4 t^4} \end{aligned} \quad (30)$$

where VXYZ are adjustable coefficients

If:

$$V = b_1 / C_1 \quad (31)$$

$$X = b_2 / D_2 \quad (32)$$

$$Y = b_3 / E_3 \quad (33)$$

$$Z = b_4 / F_4 \quad (34)$$

Then the isotopic ratio will be constant up to the fifth power of t . This analysis provides insights that suggest the use of coefficients to improve the constancy of isotopic ratios. The detailed theoretical discussion of this approach is in the following section.

IV. THE DEFINITION OF ISOTOPIC RATIOS AND THE LIMITS OF THEIR RELIABILITY AND APPLICABILITY

A. THE GENERAL PROCESS OF CONSTRUCTING ISOTOPIC RATIOS

The construction of isotopic ratios ideally should be based on isotopic concentrations, measured accurately, or calculated using the best available calculational methods. The development of the ratios should be carried out before the need to apply them in safeguards actually arises. This approach ensures that the techniques will be thoroughly explored before they are needed in applications. This approach minimizes the probability of "surprises" in the application of the ratios, and gives the planning for the application of the ratios and the selection of experimental data for testing a firm theoretical basis.

The broad objective of the ratio construction process is to produce two things:

- The definition of the ratio itself.
- The specification of the characteristics of the ratio over a range of variation of reactor parameters that influence the production and depletion of isotopes being safeguarded.

The steps in the construction process are as follows:

- Choose isotopic concentrations that are representative of the range of application of the ratio and systematically define a ratio that promises to have the desired accuracy and other desired properties over the range of application.
- Determine the deviation of the ratio from its desired characteristics over the range of application using isotopic concentrations calculated with the best available calculational methods and experimental data if they exist.

- If the deviation of the ratio from its desired characteristics is too large or the range of application too limited, then apply ratio range and accuracy extension methods as described in the examples given later.
- Verify the new range and accuracy of the ratio by performing the second step again.

The first step in the construction process speaks of "systematically defining a ratio." This systematic construction process is one that can be accomplished by individuals with understanding and experience in the field of isotopic ratios. It depends on a familiarity with the functional forms of the various isotopic concentrations as a function of fuel exposure and how these forms should be blended to produce desired properties such as constancy as a function of exposure or moderator void. The purpose of the second step in the construction process is to determine the accuracy and reliability of an isotopic ratio over a specified range of application. However, the accuracy of the determination depends to some extent on the accuracy of the calculational methods used to calculate the isotopic concentrations. The expected accuracy of isotopic measurement in practice should also be considered in choosing among alternate ratios. Reference 5 discusses the accuracy of calculational methods in detail. The conclusions of Reference 5 with regard to the accuracy of calculational methods are summarized in Table 1.

The best accuracy currently achieved in calculating the plutonium isotopic concentrations in irradiated fuels is 5% for ^{239}Pu . The poorest accuracy is 15% for ^{242}Pu . These accuracy levels point out the need for experimental data to properly normalize isotopic ratios for safeguards applications. In spite of these limitations, calculations are still useful in the following applications:

- Direct safeguards applications in situations where no experimental information exists. Knowledge of the isotopic ratio with its limitations is better than no knowledge at all.

TABLE 1

CURRENT AND PROBABLE FUTURE UNCERTAINTIES IN PREDICTED
ISOTOPIC CONTENT OF POWER REACTOR CORES

<u>Parameter</u>	<u>Probable Future Capability**</u>	<u>Current Performance*</u>
^{235}U	1.5%	3%
^{236}U	3%	6%
^{238}U	0.1%	0.1%
Total U	0.1%	0.1%
^{239}Pu	3%	5%
^{240}Pu	6%	8%
^{241}Pu	9%	11%
^{242}Pu	11%	15%
Total Pu	3%	6%

* Assuming $\pm 10\%$ error in thermal power.

** Assuming factor of two increase in accuracy of thermal power measurement.

- Construction and testing of isotopic ratios, especially where insufficient experimental data exists to do the job. Calculated isotopic concentrations permit the development and investigation of isotopic ratios for future applications for which there is not yet experimental data. In this way the very first sets of experimental data to be obtained can be handled with experience and with methods that are already highly developed.

The following are examples of calculational methods available at BNW which can be applied to the calculation of isotopic concentrations.

HRBRT Unified thermal and fast neutron spectrum calculation for reactors using revised versions of THERMOS and GAM-I.

HAMMER	One-dimensional heterogeneous thermal and fast spectrum calculation using revised versions of THERMOS and MUFT with a one dimensional diffusion calculation.
DTF-IV	Solves the one dimensional time-independent Boltzman equation for neutron distributions as a function of energy, space and angular dependence.
LASER	Calculates the neutron spectrum in a uniform lattice (fuel and moderator regions) with cylindrical geometry using THERMOS and MUFT.
LEOPARD	Unit cell homogenization and spectrum generation program with a fuel depletion option.
TWODB	Two-dimensional multigeometry diffusion theory program for reactor analysis and depletion.
PDQ-5	Solves the few group two-dimensional equations in multigeometry with a depletion option.
TRIDB	Three-dimensional multigeometry diffusion theory program for reactor analysis.
ALTHAEA	One-dimensional reactor depletion calculation using an approximate spectrum calculation for scoping studies.
ZODIAC	One-dimensional diffusion and depletion calculation using a detailed spectrum calculation (HRBRT).
AFLARE	New version of FLARE for reactor simulation in core reactivity and power distribution studies.
LEARN	General least squares fitting routine that can be used with nonlinear fitting parameters (prepares AFLARE input).
BMC	Two- and three-dimensional MONTE CARLO Code for use in making "exact" calculations of fuel assembly power distributions.
GGC-4	Unified thermal and fast spectrum calculation for reactor using revised versions of TEMPEST and GAM-II.

B. QUALITIES OF AN APPROACH FOR CONSTRUCTING ISOTOPIC RATIOS FOR SAFEGUARDS APPLICATIONS

In a previous section the simplification approach was shown to have shortcomings with regard to accuracy and generality. In defining a practical approach to constructing isotopic ratios one must insure that the approach is general and at the same time produces ratios with a high degree of accuracy and reliability. The more the approach has these qualities the greater the probability that it will be successful in dealing with the demands of the present and future safeguards isotopic ratio requirements. Any successful approach must take into account an important quality of an isotopic ratio: its "diversion sensitivity." Diversion sensitivity is the response of a ratio to an attempted diversion. It is a measure of the reliability of a ratio in detecting diversion. Suppose that a certain amount of plutonium was diverted leaving a fraction f of the actual amount of plutonium. If the original amount of plutonium is Pu then the amount remaining is fPu , and the amount diverted is $(1-f)Pu$. In this case each of the individual plutonium isotopic concentrations will be diverted in the same proportion or:

$$fPu = f {}^{239}Pu + f {}^{240}Pu + f {}^{241}Pu + f {}^{242}Pu \quad (35)$$

This kind of diversion is defined to be proportional diversion. On the other hand the diversion may take place in a manner which also changes the relative proportions of the individual plutonium isotopes (for example by substituting plutonium low in fissile isotopic content from high burnup fuel for plutonium high in fissile isotopic content) then:

$$aPu = b {}^{239}Pu + c {}^{240}Pu + d {}^{241}Pu + e {}^{242}Pu \quad (36)$$

where: a, b, c, d and e are positive fractions. This diversion is defined to be nonproportional diversion. Some ratios such as $Pu/U/{}^{235}D$ are sensitive to proportional diversion and to some but not all, nonproportional diversion. Some ratios such as ${}^{239}Pu \cdot {}^{242}Pu / {}^{240}Pu \cdot {}^{241}Pu$ are insensitive to proportional diversion but sensitive to some types of nonproportional diversion.

A mathematical definition of diversion sensitivity is as follows.

$$S = \left[\frac{\partial R(p)}{\partial p} \right] \frac{1}{R(1)} \quad (37)$$

where: $p = f$ in defining the sensitivity to proportional diversion
or

$p = b, c, d, e$ in defining the sensitivity of individual
Pu isotopes to nonproportional diversion.

S = sensitivity to Pu diversion

$R(p)$ = an isotopic ratio which is a function of the parameter p .

As an example consider the isotopic ratio $Pu/U/U^{235}D$. If each Pu isotopic concentration in the ratio is replaced by f times the Pu isotopic concentration or:

$$\frac{f \text{ } ^{239}\text{Pu} + f \text{ } ^{240}\text{Pu} + f \text{ } ^{241}\text{Pu} + f \text{ } ^{242}\text{Pu}}{U(^{235}D)} = f \left[\frac{Pu}{U(^{235}D)} \right] \quad (38)$$

the diversion is propagated linearly into the ratio. In other words, a diversion of 10% of the material will result in a 10% decrease in the ratio. Looking at Equation (36) we can see that it is possible to choose values of b, c, d and e such that $a = 1$ (total plutonium remains constant) and yet fissile material could be diverted. In other words, some of the ^{239}Pu and ^{241}Pu could be diverted and replaced with nonfissile ^{240}Pu and ^{242}Pu in such a way that the total plutonium is unchanged. In this case, the ratio $Pu/U/U^{235}D$ would not detect the diversion. Another ratio would have to be used as a backup to detect this kind of situation.

The requirements for a suitable single backup ratio would be that the ratio be sensitive to all nonproportional diversions that satisfy Equation (36) with $a = 1$. If the backup ratio has some blind spots in ranges of interest a second backup ratio could be used to cover these blind spots and so on.

The search for the blind spots in an isotopic ratio or systems of mutually supporting isotopic ratios can be attempted by searching for solutions to a system of equations. The equations in the system

are valid only for small perturbations in the isotopics; however, diversion is likely to involve small fractions of the isotopics in order to avoid detection.

Let us assume that a diverter may attempt to divert plutonium and may attempt to mask the diversion by adjusting the ^{235}U concentration. Let us also assume that the system of independent backup ratios R_1, R_2, R_3, R_4 and R_5 contains $^{239}\text{Pu}, ^{240}\text{Pu}, ^{241}\text{Pu}, ^{242}\text{Pu}$, and $^{238}\text{U}, ^{235}\text{U}, ^{236}\text{U}$. If the diversion is to be undetected by isotopic ratios, it must produce no changes in these ratios. The conditions that the diverted isotopes must therefore satisfy are as follows:

$$\begin{aligned} \frac{\partial R_i}{\partial ^{239}\text{Pu}} \left(\Delta ^{239}\text{Pu} \right) + \frac{\partial R_i}{\partial ^{240}\text{Pu}} \left(\Delta ^{240}\text{Pu} \right) + \frac{\partial R_i}{\partial ^{241}\text{Pu}} \left(\Delta ^{241}\text{Pu} \right) \\ + \frac{\partial R_i}{\partial ^{242}\text{Pu}} \left(\Delta ^{242}\text{Pu} \right) + \frac{\partial R_i}{\partial ^{235}\text{U}} \left(\Delta ^{235}\text{U} \right) = 0 \end{aligned} \quad (39)$$

where:

$$i = 1, 2, 3, 4, 5 \dots$$

$\Delta\text{Pu}, \Delta\text{U}$ are small changes in Pu and U isotopic concentrations due to diversion.

For each backup ratio R_i there is an equation expressing the conditions under which the diversion of plutonium isotopics will produce no change in the ratio.

If 5 backup ratios are used, System (39) is a linear homogeneous system in 5 equations and 5 unknowns ($\Delta ^{239}\text{Pu}, \Delta ^{240}\text{Pu}, \Delta ^{241}\text{Pu}, \Delta ^{242}\text{Pu}, \Delta ^{235}\text{U}$). The theory of linear systems guarantees that a non-zero solution for the system exists if and only if the determinant of the partial derivative coefficients equals zero. The determinant is easily computed by standard methods. In most cases the determinant will not equal zero and no solution or blind spot will exist. If the determinant equals zero, then a sixth backup ratio can be included creating a linear system with 6 equations and 5 unknowns which theory guarantees has no

solutions or blind spots. When 4 or fewer backup ratios are used, the system is under-determined. There may be an infinite number of solutions or blind spots in the system of equations. When 4 backup ratios are used, the potential diverter can specify the ^{239}Pu or ^{241}Pu to be diverted as a free parameter and solve the resulting inhomogeneous 4×4 linear system for the other Δ s which will mask the diversion. Theory guarantees a solution if the determinant of the coefficient matrix is non-zero.

In practice, fewer than 4 backup ratios may be sufficient for application because other conditions eliminate many of the possible solutions. For example, the practical limits on the availability of certain isotopes may preclude some theoretically possible diversion schemes. In addition, changes in an isotope of a greater percentage than some nominal amount such as 30 percent would be obvious and therefore only solutions or blind spots in the range ± 30 percent are of interest. Two or three mutually supporting ratios may be practically impossible to defeat when the potential diverter does not know what ratios are being used to check him and therefore does not know how to define relationships which will tell him where the blind spots are.

The real significance of this discussion is that it points out how to design systems of isotopic ratios that mathematically guarantee against defeat by diversions or substitutions involving small perturbations in the isotopic concentrations. Changes in the isotopic concentrations greater than 30 percent should be detectable by trained inspection. Further studies will be needed to define the systems of backup ratios to be used in specific safeguards applications based on these mathematical criteria. Future studies should also determine just how large the realm of small perturbations is, and ensure that the realm of small perturbations overlaps the realm of deviations in the isotopic concentrations that are obvious to trained inspection. The necessary ingredient required in these systems of isotopic ratios is an abundance of ratios available for application. The number of isotopic ratios available from measurement data does not seem to be a limitation. The use of isotopic powers and coefficients further increases the number of suitable isotopic ratios beyond what is available without the use of coefficients, (see Example 3, Page 41).

C. EXTENDING THE RANGE AND ACCURACY OF ISOTOPIC RATIOS

Isotopic ratios are useful for safeguards applications because they extend the intercomparability of isotopic data. In the absence of the ratio $\text{Pu}/\text{U}/^{235}\text{D}$ or a ratio like it, only isotopic data carefully matched in exposure and all the other variables that affect the net plutonium production could be compared for consistency. The $\text{Pu}/\text{U}/^{235}\text{D}$ ratio provides a basis for comparison over a wide range of exposure for certain types of reactor data. Similarly the ratio $[(^{240}\text{Pu})^2/(^{239}\text{Pu} \cdot ^{242}\text{Pu})] [\text{Pu}/\text{U}]$ provides a basis for comparison over ranges of exposure and moderator void. These characteristics are particularly useful in the analysis of BWR's.

Neither of these ratios, however, provide a perfect basis of comparison. That is, neither of them are perfectly constant as a function of exposure or void. Both of them vary as a function of exposure and void, but the variation is small enough so that the ratio is useful in spite of it. Both of these ratios could be improved by reducing this background variation. Actually, a comparison of the sensitivity of a ratio with its background variation in percent is the best indicator of the usefulness and reliability of a ratio since the ratio should move significantly outside of the band of background variations in response to a significant diversion. The methods of extending the range and accuracy of isotopic ratios do so by decreasing the background variations while maintaining an adequate level of sensitivity.

The previous section presented the idea of using coefficients to multiply the Pu isotopic concentrations in order to achieve improved accuracy and range of applicability in isotopic ratios. This is one of two basic approaches that can be used. The two approaches are:

- The ratio has certain terms which are the sums of isotopic concentrations. In this case isotopic concentrations in the sums are multiplied by coefficients which reduces the background variations of the isotopic ratio. Example: The ratio

$$\frac{^{239}\text{Pu} + ^{241}\text{Pu}}{^{242}\text{Pu}} \quad \text{becomes} \quad \frac{^{239}\text{XPu} + ^{241}\text{YPu}}{^{242}\text{Pu}}$$

where X and Y are coefficients.

- The ratio has certain parts which are composed of products and quotients of isotopic concentrations. The isotopic concentrations in these products and quotients are raised to powers which reduce the background variations of the isotopic ratio.

Example: the ratio

$$\frac{{}^{239}\text{Pu} \quad {}^{242}\text{Pu}}{{}^{240}\text{Pu} \quad {}^{241}\text{Pu}} \quad \text{becomes} \quad \frac{({}^{239}\text{Pu})^X ({}^{242}\text{Pu})^Y}{({}^{240}\text{Pu})^W ({}^{241}\text{Pu})^Z}$$

where X, Y, W and Z are powers.

In actual applications either approach or a combination of the approaches may be used.

The following example illustrates that the use of isotopic powers is equivalent to having a limited capacity to adjust the basic nuclear cross sections in order to obtain constancy in an isotopic ratio.

Example 1. The Relationship Between Isotopic Powers and Effective Cross Sections

Let us assume that in constructing a ratio with a high degree of constancy one chooses isotopes A and B such that:

- $N^A(t)$ satisfies the transmutation equation:

$$\frac{dN^A(t)}{dt} = - N^A(t) \bar{\sigma}_a^A(t) \phi(t) \quad . \quad (40)$$

- $N^B(t)$ satisfies the transmutation equation:

$$\frac{dN^B(t)}{dt} = - N^B(t) \bar{\sigma}_a^B(t) \phi(t) \quad . \quad (41)$$

The solutions to the transmutation equations are as follows:

$$N^A(t_0) = N^A(0) e^{-\int_0^{t_0} \bar{\sigma}_a^A(t) \phi(t) dt} \quad . \quad (42)$$

$$N^B(t_0) = N^B(0) e^{-\int_0^{t_0} \bar{\sigma}_a^B(t) \phi(t) dt} \quad . \quad (43)$$

The solutions (42) and (43) can be used to form the isotopic ratio:

$$\frac{N^A(t_0)}{N^B(t_0)} = \frac{N^A(o)e^{-\int_0^{t_0} \bar{\sigma}_a^A(t)\phi(t)dt}}{N^B(o)e^{-\int_0^{t_0} \bar{\sigma}_a^B(t)\phi(t)dt}} \quad (44)$$

or

$$\frac{N^A(t_0)}{N^B(t_0)} = \frac{N^A(o)}{N^B(o)} \left[e^{-\int_0^{t_0} \phi(t) \left(\bar{\sigma}_a^A(t) - \bar{\sigma}_a^B(t) \right) dt} \right] \quad (45)$$

The isotopic ratio expressed in (45) will be constant if

$$\int_0^{t_0} \phi(t) \left(\bar{\sigma}_a^A(t) - \bar{\sigma}_a^B(t) \right) dt = C \quad (46)$$

where C is a constant.

In fact, the search for isotopes A and B such that the isotopic ratio $N^A(t)/N^B(t)$ as closely as possible equals the initial value $N^A(o)/N^B(o)$ over the whole exposure range is equivalent to searching for isotopes with effective absorption cross sections that satisfy the condition:

$$\int_0^{t_0} \phi(t) \left(\bar{\sigma}_a^A(t) - \bar{\sigma}_a^B(t) \right) dt = 0 \quad (47)$$

Usually the selection of isotopes available does not contain a pair with the precisely correct requirements. The background variations in an isotopic ratio are in part due to the mismatch in the effective cross sections.

This limitation can be overcome in part by raising the isotopes in the ratio to appropriate powers. Raising both sides of (42) to the power X we have:

$$\left[N^A(t_0) \right]^X = \left[N^A(o) \right]^X e^{-\int_0^{t_0} X \bar{\sigma}_a^A(t) \phi(t) dt} \quad (48)$$

Raising both sides of (43) to the power Y we have:

$$\left[N^B(t_0) \right]^Y = \left[N^B(0) \right]^Y e^{-\int_0^{t_0} Y \bar{\sigma}_a^B(t) \phi(t) dt} \quad (49)$$

Equation (45) with the powers becomes

$$\frac{\left[N^A(t_0) \right]^X}{\left[N^B(t_0) \right]^Y} = \frac{\left[N^A(0) \right]^X}{\left[N^B(0) \right]^Y} \left[e^{-\int_0^{t_0} \phi(t) \left(X \bar{\sigma}_a^A(t) - Y \bar{\sigma}_a^B(t) \right) dt} \right] \quad (50)$$

The condition that the isotopic ratio equal the initial value over the whole exposure ranges is

$$\int_0^{t_0} \phi(t) \left(X \bar{\sigma}_a^A(t) - Y \bar{\sigma}_a^B(t) \right) dt = 0 \quad (51)$$

It can be seen that the introduction of the powers X and Y in the ratio is equivalent to multiplying the effective cross section by the values of the powers. The values of the powers may be chosen such that the expression (51) is satisfied as closely as possible in the exposure range. When (51) is satisfied, Equation (50) reduces to the form of (52) and (53). Equations (52) and (53) require that the value of the ratio at exposure times t_2 and t_1 equal the value of the ratio at the initial time $t = 0$. Equations (52) and (53) are used to solve for X and Y.

$$\frac{\left[N^A(t_1) \right]^X}{\left[N^B(t_1) \right]^Y} = \frac{\left[N^A(0) \right]^X}{\left[N^B(0) \right]^Y} \quad (52)$$

$$\frac{\left[N^A(t_2) \right]^X}{\left[N^B(t_2) \right]^Y} = \frac{\left[N^A(0) \right]^X}{\left[N^B(0) \right]^Y} \quad (53)$$

The basic approach illustrated in this example has been applied to a variety of isotopic ratios that have safeguards applications. The ratios have been composed of products, sums and quotients of isotopics.

Similar derivations have been successfully performed in all these situations. The following examples illustrate various applications of these techniques.

Example 2: A Linear System that Reduces the Background Variation in Pu/U/²³⁵D as Applied to YANKEE ROWE Data

From Figure 9 in the Appendix, it can be seen that in the exposure range 7500 to 25,000 MWd/MTU, Pu/U/²³⁵D varies about 3% when applied to YANKEE ROWE data. This background variation reduces the ability of the ratio to detect diversions of less than 3% in Pu. The purpose of this example is to demonstrate the use of isotope coefficients in reducing the background variation in the isotopic ratio Pu/U/²³⁵D and to show the effect of coefficients on diversion sensitivity and on the ratio of diversion sensitivity to percent background variation. The following linear system in six equations and six unknowns defines coefficients that require the isotopic ratio to equal a chosen low exposure value of the ratio at six points in the exposure range:

$$\left[\frac{1}{^{235}\text{D}_{(i)}} \right] \left[\frac{A \text{ } ^{239}\text{Pu}_{(i)} + B \text{ } ^{240}\text{Pu}_{(i)} + C \text{ } ^{241}\text{Pu}_{(i)} + D \text{ } ^{242}\text{Pu}_{(i)}}{^{238}\text{U}_{(i)} + F \text{ } ^{236}\text{U}_{(i)} + G \text{ } ^{235}\text{U}_{(i)}} \right] =$$

$$\left[\frac{1}{^{235}\text{D}_{(0)}} \right] \left[\frac{^{239}\text{Pu}_{(0)} + ^{240}\text{Pu}_{(0)} + ^{241}\text{Pu}_{(0)} + ^{242}\text{Pu}_{(0)}}{^{238}\text{U}_{(0)} + ^{236}\text{U}_{(0)} + ^{235}\text{U}_{(0)}} \right]$$

$$i = 1, 2, 3, 4, 5, 6 \quad (54)$$

Where: A, B, C, D, F, G are coefficients to be determined.

Pu_(i), U_(i) are isotopic concentrations at exposure points i (i = 0 is the initial point to which all the other points are set equal). System (54) can be rearranged into the familiar form of a linear 6 x 6 system.

$$\begin{aligned}
& A \text{ } ^{239}\text{Pu}_{(i)} + B \text{ } ^{240}\text{Pu}_{(i)} + C \text{ } ^{241}\text{Pu}_{(i)} + D \text{ } ^{242}\text{Pu}_{(i)} - F \left[\frac{C_0 \text{ } ^{236}\text{U}_{(i)}}{R_{(i)}} \right] \\
& - G \left[\frac{C_0 \text{ } ^{235}\text{U}_{(i)}}{R_{(i)}} \right] = \frac{C_0 \text{ } ^{238}\text{U}_{(i)}}{R_{(i)}} \quad (55)
\end{aligned}$$

where:

$$\begin{aligned}
C_0 &= \left[\frac{1}{^{235}\text{D}_{(0)}} \right] \left[\frac{^{239}\text{Pu}_{(0)} + ^{240}\text{Pu}_{(0)} + ^{241}\text{Pu}_{(0)} + ^{242}\text{Pu}_{(0)}}{^{238}\text{U}_{(0)} + ^{236}\text{U}_{(0)} + ^{235}\text{U}_{(0)}} \right] \\
R_{(i)} &= \frac{1}{^{235}\text{D}_{(i)}} \quad i = 1, 2, 3, 4, 5, 6
\end{aligned}$$

There are substantial advantages in obtaining coefficients by defining a linear system as opposed to defining a non-linear system. If the determinant of an inhomogeneous linear system is non zero, a solution exists. If a solution exists, there are standard methods of calculating the solution which do not require prior knowledge of starting values. The solution of the linear system for the exposure points listed in Table II is as follows:

$$\begin{aligned}
A &= 0.3254 \\
B &= 0.7038 \\
C &= 0.5690 \\
D &= 1.504 \\
F &= -18.36 \\
G &= -64.66
\end{aligned} \quad (56)$$

The effect of these coefficients on the background variation of the isotopic ratio is also shown in Table II.

TABLE II
A COMPARISON OF THE BACKGROUND
VARIATIONS OF Pu/U/²³⁵D WITH AND WITHOUT
COEFFICIENTS USING YANKEE ROWE CALCULATED DATA

<u>Points used in Linear System</u>	<u>Exposure Time in Days</u>	<u>Exposure in Mwd/T</u>	<u>Percent Variation with Coefficients</u>	<u>Percent Variation without Coefficients</u>
0	40	1128	- 0.0720	- 0.0
1	100	2820	- 0.0074	- 1.03
2	200	5640	- 0.0043	- 1.75
	300	8460	- 0.00033	- 1.89
3	400	11280	- 0.0035	- 1.78
	500	14100	- 0.0035	- 1.50
4	600	16920	- 0.0028	- 1.10
	700	19740	- 0.0031	- 0.66
5	800	22560	- 0.0024	- 0.0088
	900	25380	- 0.0050	+ 0.68
6	925	26085	- 0.0022	+ 0.26

Table II compares the percent variation of the isotopic ratio Pu/U/²³⁵D as a function of exposure with and without the use of coefficients. The variations of exposure points used in defining the linear system as well as some intermediate points are compared. The intermediate points are included to show that the coefficients reduce the variation at all points to a similar degree.

It can be seen that the variation has a range of about 3% (rounded to the nearest percent) when coefficients are not used while the variation is reduced by roughly a factor of 1000 when coefficients are used

over the exposure range of practical application from 2,820 to 26,085 MWd/MTU. One cannot conclude from the table that the use of coefficients alone makes it possible to safeguard Pu isotopes to within thousandths of a percent using isotopic ratios. Typical measurement errors of the individual isotopic concentrations are much larger than this ranging from tenths of a percent for ^{239}Pu and ^{238}U to several percent for ^{242}Pu and ^{236}U . These errors are the limiting factor in the accuracy attainable for isotopic ratios. The table does show, however, that the use of coefficients can eliminate background variations as a limiting factor in the constancy of isotopic ratios, so that as the accuracy of isotopic concentration measurements increases it will be possible to exploit fully these improvements in accuracy in the application of isotopic ratios to safeguards.

Decreasing the background variations alone does not insure improved performance of an isotopic ratio in safeguards applications. A ratio with safeguards applications should change significantly more in response to diversion than the magnitude of its background variations. For this reason the ratio of sensitivity to the percent background variation is the practical measure of isotopic ratio performance. The sensitivity of the ratio $\text{Pu}/\text{U}/^{235}\text{D}$ to proportional diversion of Pu (no other isotopes are changed) is the same with or without coefficients. The sensitivity of the ratio $\text{Pu}/\text{U}/^{235}\text{D}$ to nonproportional diversion may increase or decrease when coefficients are used depending upon which Pu isotopes are altered by the diversion. Thus, for diversion which alters the ^{239}Pu concentration the calculated sensitivity for the $\text{Pu}/\text{U}/^{235}\text{D}$ ratio with coefficients using data at 11,280 MWd/MTU is 18 percent lower than the sensitivity of the ratio without coefficients. For diversions involving ^{242}Pu changes, the calculated sensitivity of the ratio with coefficients using data at 11,280 MWd/MTU is 2.8 times larger than the sensitivity of the ratio without coefficients. For either of these isotopes the 1000 fold reduction in background variation obtained by using coefficients will easily cause a net increase in the ratio of sensitivity to background by at least a factor of 100 causing a substantial increase in the potential usefulness and reliability of the isotopic ratios for safeguards purposes.

Example 3: Linear Systems that Reduce the Background Variation and Increase Reliability of the Ratio $Pu/(^{235}U + ^{236}U)$ as Applied to the Saxton Pu Recycle Core

It is likely that there will be a significant number of plutonium enriched light water reactor cores that will be built, irradiated and processed in the future. It will be as useful to safeguard the plutonium in these cores using isotopic ratios as it is in uranium enriched cores. An important question which must be answered is what isotopic ratios can be used to safeguard Pu enriched cores. The work on the complete answer to this question is still in progress. The work so far based on isotopic concentrations from the Saxton Pu recycle core indicates no lack of potential for success in this area. The ratio $Pu/(^{235}U + ^{236}U)$ applied to Saxton has about the same magnitude of background variation as a function of exposure as $Pu/U/^{235}D$ applied to YANKEE ROWE. This ratio is a candidate for safeguards applications to Pu recycle cores. However, its range of application must yet be determined by applying it in a wide range of Pu recycle irradiation conditions.

The purpose of this example is:

- To demonstrate the correction of the background variations in the ratio $Pu/(^{235}U + ^{236}U)$ using two different linear systems which produce two distinct sets of solution coefficients.
- To demonstrate the use of multiple sets of coefficients to build a system of backup ratios with coefficients from a single ratio, thus creating a new dimension of difficulty for the diverter who is searching for blind spots in the isotopic ratios.

The first system of equations has 3 equations and 3 coefficients:

$$\frac{A \ ^{239}Pu_{(i)} + B \ ^{240}Pu_{(i)} + C \ ^{241}Pu_{(i)} + ^{242}Pu_{(i)}}{^{235}U_{(i)} + ^{236}U_{(i)}} = \frac{A \ ^{239}Pu_{(0)} + B \ ^{240}Pu_{(0)} + C \ ^{241}Pu_{(0)} + ^{242}Pu_{(0)}}{^{235}U_{(0)} + ^{236}U_{(0)}} \quad i = 1, 2, 3, \quad (57)$$

Where: A, B, C, are coefficients to be determined

$Pu_{(i)}$, $U_{(i)}$ are isotopic concentrations at exposure times i ,
($i = 0$ is the initial point to which all the other points are
set equal)

System (57) can be rearranged into the standard form of a 3×3 system
as follows:

$$\begin{bmatrix} W_1 & X_1 & Y_1 \\ W_2 & X_2 & Y_2 \\ W_3 & X_3 & Y_3 \end{bmatrix} \begin{bmatrix} A \\ B \\ C \end{bmatrix} = \begin{bmatrix} Z_1 \\ Z_2 \\ Z_3 \end{bmatrix}$$

where:

$$\begin{aligned} W_i &= {}^{239}\text{Pu}_{(i)} ({}^{235}\text{U}_{(0)} + {}^{236}\text{U}_{(0)}) - {}^{239}\text{Pu}_{(0)} ({}^{235}\text{U}_{(i)} + {}^{236}\text{U}_{(i)}) \\ X_i &= {}^{240}\text{Pu}_{(i)} ({}^{235}\text{U}_{(0)} + {}^{236}\text{U}_{(0)}) - {}^{240}\text{Pu}_{(0)} ({}^{235}\text{U}_{(i)} + {}^{236}\text{U}_{(i)}) \\ Y_i &= {}^{241}\text{Pu}_{(i)} ({}^{235}\text{U}_{(0)} + {}^{236}\text{U}_{(0)}) - {}^{241}\text{Pu}_{(0)} ({}^{235}\text{U}_{(i)} + {}^{236}\text{U}_{(i)}) \\ Z_i &= {}^{242}\text{Pu}_{(0)} ({}^{235}\text{U}_{(i)} + {}^{236}\text{U}_{(i)}) - {}^{242}\text{Pu}_{(i)} ({}^{235}\text{U}_{(0)} + {}^{236}\text{U}_{(0)}) \end{aligned}$$

$i = 1, 2, 3. \quad (58)$

The second system of equations has 5 equations and 5 coefficients:

$$\begin{aligned} & \frac{A {}^{239}\text{Pu}_{(i)} + B {}^{240}\text{Pu}_{(i)} + C {}^{241}\text{Pu}_{(i)} + D {}^{242}\text{Pu}_{(i)}}{E {}^{235}\text{U}_{(i)} + {}^{236}\text{U}_{(i)}} \\ &= \frac{{}^{239}\text{Pu}_{(0)} + {}^{240}\text{Pu}_{(0)} + {}^{241}\text{Pu}_{(0)} + {}^{242}\text{Pu}_{(0)}}{{}^{235}\text{U}_{(0)} + {}^{236}\text{U}_{(0)}} \end{aligned}$$

$$i = 1, 2, 3, 4, 5 \quad (59)$$

System (59) can be rearranged into the standard form as follows:

$$A \text{ } ^{239}\text{Pu}_{(i)} + B \text{ } ^{240}\text{Pu}_{(i)} + C \text{ } ^{241}\text{Pu}_{(i)} + D \text{ } ^{242}\text{Pu}_{(i)} - E \text{ } ^{235}\text{C}_0\text{U}_{(i)} = \text{ } ^{236}\text{C}_0\text{U}_{(i)} \quad (60)$$

where:

$$C_0 = \frac{^{239}\text{Pu}_{(0)} + ^{240}\text{Pu}_{(0)} + ^{241}\text{Pu}_{(0)} + ^{242}\text{Pu}_{(0)}}{^{235}\text{U}_{(0)} + ^{236}\text{U}_{(0)}}$$

The solution of the 3 x 3 system for the Saxon fuel is as follows:

$$\begin{aligned} A &= -1.312 \\ B &= -1.501 \\ C &= -0.7016 \end{aligned} \quad (61)$$

The solution of the 5 x 5 system is:

$$\begin{aligned} A &= 0.5680 \\ B &= 0.8008 \\ C &= 0.4064 \\ D &= 0.05151 \\ E &= 0.5864 \end{aligned} \quad (62)$$

The effect of these coefficients on the background variation of the isotopic ratio is shown in Table III.

The maximum percent variation of the isotopic ratio without coefficients shown in Table III is 3.6 percent. Typical variations when either 3 or 5 coefficients are used are about a factor of a thousand smaller so that they do the job of reducing background variations about equally well.

TABLE III

A COMPARISON OF THE BACKGROUND VARIATIONS
OF Pu/(²³⁵U + ²³⁶U) WITH AND WITHOUT COEFFICIENTS
USING SAXTON Pu RECYCLE DATA

<u>Points Used in Linear System</u>	<u>Exposure in Mwd/MTU</u>	<u>% Variation without Coefficients</u>	<u>% Variation with 3 Coefficients</u>	<u>% Variation with 5 Coefficients</u>
0 X	0.0	0.0	0.0	0.0
X	2414.5	+ 0.140	+ 0.0045	+ 0.007
X	7108.5	+ 0.480	+ 0.0067	+ 0.0074
0	9455.7	+ 0.690	+ 0.0069	
	11803.	+ 0.936	+ 0.00084	+ 0.001
X	14150.5	+ 1.22		+ 0.007
	16498.	+ 1.53	+ 0.00034	+ 0.0014
0	18846.	+ 1.88	+ 0.000003	
X	21194.	+ 2.26	+ 0.0033	+ 0.0073
	25890.8	+ 3.13	+ 0.003	+ 0.009
0 X	28239.6	+ 3.62	+ 0.0	+ 0.008

0: identifies points used in the 3 x 3 system

X: identifies points used in the 5 x 5 system

An important advantage in solving the 3 and the 5 coefficient formulation is that the coefficients transform the single original ratio into a system of two mutually supporting ratios. We can see that the $Pu/(^{235}U + ^{236}U)$ ratio has the same kind of blind spot as was pointed out in the ratio $Pu/U/^{235}D$. If only the one ratio $Pu/(^{235}U + ^{236}U)$ without coefficients were available to safeguard Pu, the potential diverter could calculate the kinds of diversion that would not be detected by the ratio. The two ratio system is much more difficult to defeat. The introduction of coefficients into the ratio adds a new dimension in the scope of the application of the correlations for the safeguarding agency and a new dimension of difficulty for the diverter attempting to defeat the ratios.

Example 4: Reducing the Background Variation in

$$\frac{(^{240}Pu)^2}{^{239}Pu \cdot ^{242}Pu} \left[\frac{Pu}{U} \right]$$

For a BWR

The BWR can present difficulties not found in the PWR regarding the application of isotopic ratios to safeguards. The steam bubbles in the BWR change the effective moderator-to-fuel ratio in the reactor and this in turn has a significant effect on the burnup paths of the plutonium isotope. The steam fraction in the BWR reactor core depends on the reactor power level. In a reactor generating electricity the power level follows the electrical load being placed on the power plant. In the BWR situation an isotopic ratio that has constancy as a function of moderator void fraction and exposure is necessary. The ratio $Pu/U/^{235}D$ has more variation than desired as a function of void fraction. The ratio

$$\frac{(^{240}Pu)^2}{(^{239}Pu \cdot ^{242}Pu)} \left[\frac{Pu}{U} \right]$$

has less variation as a function of void fraction and exposure and

from the standpoint of background variations is better suited for application to BWR fuel. The purpose of this example is:

- To demonstrate the use of coefficients to reduce the background variations as a function of void and exposure in the isotopic ratio

$$\frac{({}^{240}\text{Pu})^2}{({}^{239}\text{Pu} \cdot {}^{242}\text{Pu})} \left[\frac{\text{Pu}}{\text{U}} \right]$$

- To demonstrate the effectiveness of linear and nonlinear systems in reducing large background variations. In previous examples the background variations of the ratios without coefficients are less than 4 percent. In this example the background variation without coefficients is as much as 34 percent.

The first system of equations is nonlinear. It has 4 equations and 4 coefficients.

$$\begin{aligned} R(i) & \left[\frac{A {}^{239}\text{Pu}(i) + B {}^{240}\text{Pu}(i) + {}^{241}\text{Pu}(i) + {}^{242}\text{Pu}(i)}{E {}^{238}\text{U}(i) + F {}^{235}\text{U}(i) + {}^{236}\text{U}(i)} \right] \\ &= R(0) \left[\frac{A {}^{239}\text{Pu}(0) + B {}^{240}\text{Pu}(0) + {}^{241}\text{Pu}(0) + {}^{242}\text{Pu}(0)}{E {}^{238}\text{U}(0) + F {}^{235}\text{U}(0) + {}^{236}\text{U}(0)} \right] \\ & \quad i = 1, 2, 3 \end{aligned} \tag{63}$$

where: A, B, E, F are coefficients to be determined

$\text{Pu}(i)$, $\text{U}(i)$ are isotopic concentrations at exposure points i ,

($i = 0$ is the initial point to which all other points are set equal)

$$R(i) = ({}^{240}\text{Pu}(i))^2 / ({}^{239}\text{Pu}(i) \cdot {}^{242}\text{Pu}(i)) \quad i = 0, 1, 2, 3$$

The second system of equations is linear and has 6 equations and 6 coefficients.

$$\begin{aligned}
 R(i) & \left[\frac{A \cdot {}^{239}\text{Pu}(i) + B \cdot {}^{240}\text{Pu}(i) + C \cdot {}^{241}\text{Pu}(i) + D \cdot {}^{242}\text{Pu}(i)}{{}^{238}\text{U}(i) + F \cdot {}^{235}\text{U}(i) + G \cdot {}^{236}\text{U}(i)} \right] \\
 & = R(o) \left[\frac{{}^{239}\text{Pu}(o) + {}^{240}\text{Pu}(o) + {}^{241}\text{Pu}(o) + {}^{242}\text{Pu}(o)}{{}^{238}\text{U}(o) + {}^{235}\text{U}(o) + {}^{236}\text{U}(o)} \right] \quad (64)
 \end{aligned}$$

This linear system is the same as that of Example 2 except for the definition of $R(i)$.

The solution of the 4 x 4 nonlinear system is as follows:

$$\begin{aligned}
 A & = - 6.479 \cdot 10^{-3} \\
 B & = 4.453 \cdot 10^{-2} \\
 E & = 7.336 \cdot 10^{-3} \\
 F & = - 2.981 \cdot 10^{-1} \quad (65)
 \end{aligned}$$

The solutions to two 6 x 6 linear systems were obtained. The differences in these two systems are in the data points used in defining the systems. Table IV shows the data points used in each of these systems labeled Y for the system defined with points from the 0 and 70 percent void data and Z for the system defined with points from the 38 and 70 percent void data.

The solution of the Y system is

$$\begin{aligned}
 A & = - 5.930 \cdot 10^{-2} \\
 B & = 1.149 \\
 C & = 5.721 \cdot 10^{-2} \\
 D & = 4.778 \\
 F & = - 37.83 \\
 G & = - 109.7 \quad (66)
 \end{aligned}$$

TABLE IV

A COMPARISON OF THE BACKGROUND VARIATIONS
 OF $[(^{240}\text{Pu})^2 / (^{239}\text{Pu} \cdot ^{242}\text{Pu})][\text{Pu}/\text{U}]$
 WITH AND WITHOUT COEFFICIENTS USING CALCULATED BWR DATA

Points Used In the Systems	Exposure in MWd/T	% Variation without Coefficients	% Variation with 4 Coefficients	% Variation** with 6 Coefficients, Y System	% Variation** with 6 Coefficients, Z System
0 Percent Void					
XY	5000	* 0.0	+26.0	- 0.945	+ 20.7
	11500	-15.6	+ 1.13	+ 0.351	+ 18.8
	13000	-17.9	- 1.06	+ 0.028	+ 18.5
	16000	-21.4	- 4.70	- 0.162	+ 19.2
Y	20500	-26.0	- 5.60	0.000	+ 22.0
	26500	-31.6	- 5.20	+ 0.113	+ 25.7
Y	28000	-32.0	- 4.90	+ 0.001	+ 26.4
38 Percent Void					
X Z	5000	-11.3	+ 28.3	- 7.80	+ 6.29
	11500	-23.3	* 0.0	- 9.80	+ 0.648
	13000	-24.7	- 1.10	-10.4	+ 0.00134
	16000	-27.0	- 4.20	-11.5	- 0.333
Z	20500	-30.0	- 2.30	-12.8	+ 0.000865
X	26500	-33.2	- 1.10	-14.6	+ 0.245
Z	28000	-34.1	- 0.600	-15.3	+ 0.000630
70 Percent Void					
X Y Z	5000	- 9.30	+ 8.20	- 3.58	+ 13.4
	11500	-11.0	0.0	- 0.0191	- 0.00972
	13000	-10.4	- 1.10	- 0.00100	+ 0.00657
	16000	- 8.80	- 2.90	- 0.0138	- 0.0553
Y Z	20500	- 5.80	- 5.50	- 0.00090	+ 0.00267
	26500	- 1.40	- 8.90	+ 0.00550	+ 0.0214
Y Z	28000	- 0.340	- 9.80	- 0.00090	+ 0.00173

* Percent deviation from this value of the isotopic ratio

** Percent deviation from $2.54115 \cdot 10^{-2}$

X: Point used in the nonlinear 4 coefficient system.

Y: Point used in the linear 6 coefficient system relating 0 and 70% void

Z: Points used in the linear 6 coefficient system relating 38% and 70% void.

The solution of the Z system is

$$A = - 0.1044$$

$$B = 1.361$$

$$C = - 0.7430$$

$$D = 6.570$$

$$F = - 39.02$$

$$G = - 127.2 \quad (67)$$

The effect of these coefficients on the background variation of the isotopic ratio is shown in Table IV. The data shown in Table IV cover the exposure range from 5,000 to 28,000 MWd/MTU. Moderator void variations are represented by 0 void, 38 percent void (a nominal average void), and 70 percent void. The maximum percent variation of the isotopic ratio without coefficients shown in Table IV is 34 percent. This is much larger than the 3 to 4 percent variations in the previous examples. This problem also has the compound variation of exposure and void that must be corrected rather than the simple variation over exposure as in the previous examples. These characteristics make this example a severe test of the coefficient method.

The 4 coefficients from the nonlinear system reduced the variation to a maximum of 10 percent in the exposure range 11,500 to 28,000 MWd/MTU. The variation is reduced to less than 1 percent by the coefficients from the 6 coefficient Y system in the exposure range 11,500 to 28,000 MWd/MTU for 0 and 70 percent voids. The variation is also reduced to less than 1 percent by the coefficients from the 6 coefficient Z system in the exposure range 11,500 to 28,000 MWd/MTU for 38 and 70 percent voids. The data points used to define these systems were chosen to ensure constancy in the middle and high exposure regions. Other coefficients can be derived that emphasize the middle and low exposure regions.

The ratios in both of these systems were set to the same numerical value so that the 0 and 38 percent void data can be indirectly compared with one another by comparing each to the 70 percent void data. It is clear that coefficients can be used to reduce large compound background variations in isotopic ratios over selected exposure and void ranges.

The previous examples have shown some applications of coefficients to reduce the background variations in isotopic ratios. These examples were chosen as strong tests of the ability of coefficients to reduce background variations. Specifically the exposure range of 0 to 26,000 MWd/MTU is larger than the largest exposure range (15,000 MWd/MTU) in any reprocessing campaign analyzed so far. The void variations given in Example 4 (0 and 70% void) are extreme variations. The average void in typical discharged BWR fuel elements will be well within this range. In addition, the examples with small background variations: 4% or less (Examples 2 and 3) had this variation reduced to thousandths of a percent. In practical applications it would only be necessary to keep the background variations below the level at which they are limiting. However, the best measurement accuracy now available on plutonium is 0.5 percent so that the background variations could perhaps be allowed to increase by a factor of 100 without being limiting. These more relaxed conditions would allow the job of reducing background variations to be done with much fewer coefficients (2 or 3) than were used in these examples. This reduction in the number of coefficients would simplify the use of coefficients and also give the coefficients a wider range of application. For example, if only 4 coefficients were being used in the isotopic ratio $\text{Pu/U}/^{235}\text{D}$, they could be applied only to the 4 plutonium isotopes thus making the coefficient set insensitive to the ^{236}U concentration. The same quality of measured data must be used in generating coefficients as was used in defining and developing isotopic ratios. Specifically, it is necessary to minimize the effects of measurement errors, and also to ensure that the data used to generate coefficients do not in themselves represent a case of diversion.

The range of applicability of a given set of coefficients must be given careful consideration. In order to gain the proper perspective on this question, it is necessary to make the observation that a set of almost universal powers and coefficients have been applied with some success in all the isotopic ratio safeguards applications that have been studied to date. In other words, the ratio "without" coefficients and powers can be considered as a "special case" of a ratio with powers and coefficients all equal to ± 1 . These coefficient values were assigned without a systematic analysis of the question of whether they are in any way optimal for safeguards applications. The principal point of the second section of this paper is that some of this universality and simplicity in coefficients can be traded for reductions in the background variations of the isotopic ratios. The best balance between accuracy and range of application and simplicity depends on the particular application. The criterion "that when a particular ratio value is applicable to sets of data the corresponding coefficients should also be applicable" or some other suitable criterion could be used as a standard in the definition of coefficients.

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APPENDIX A

DETAILS OF THE NEW DERIVATION

The discussion begins with a listing of the differential equations that govern the transmutation of the Pu isotopes.

$$\frac{dN^{239}(t)}{dt} = - N^{239}(t) \bar{\sigma}_a^{239}(t)\phi(t) + N^{238}(t) \bar{\sigma}_c^{238}(t)\phi(t) \quad (68)$$

$$\frac{dN^{240}(t)}{dt} = - N^{240}(t) \bar{\sigma}_a^{240}(t)\phi(t) + N^{239}(t) \bar{\sigma}_c^{239}(t)\phi(t) \quad (69)$$

$$\frac{dN^{241}(t)}{dt} = - N^{241}(t) \bar{\sigma}_a^{241}(t)\phi(t) + N^{240}(t) \bar{\sigma}_c^{240}(t)\phi(t) \quad (70)$$

$$\frac{dN^{242}(t)}{dt} = - N^{242}(t) \bar{\sigma}_a^{242}(t)\phi(t) + N^{241}(t) \bar{\sigma}_c^{241}(t)\phi(t) \quad (71)$$

where

$N^g(t)$ = atomic number density of isotope g as a function of time t .

$\bar{\sigma}_m^g(t)$ = effective microscopic cross section of kind m for isotope g as a function of time t ; i.e.,

$$\bar{\sigma}_m(t) = \frac{\int E \int \bar{r} \phi(E, \bar{r}, t) \sigma_m(E, \bar{r}, t) dE d\bar{r}}{\int E \int \bar{r} \phi(E, \bar{r}, t) dE d\bar{r}}$$

where

E = energy variable

$\bar{r}$ = space variable

$m = f$ (fission)

$m = a$ (absorption)

$m = c$ (capture)

$\phi(t)$ = neutron flux as a function of time t .

Differential Equations (68) through (71) may be added. The left hand side of the resulting equation is just the first derivative of the total Pu concentration, N^{Pu} . Since $\bar{\sigma}_c^{240}(t) \approx \bar{\sigma}_a^{240}(t)$ to within 0.5%, the terms on the right hand side involving $N^{240}(t)$ cancel each other out.

The resulting equation becomes:

$$\begin{aligned} \frac{dN^{Pu}(t)}{dt} = & \left[N^{238}(t) \bar{\sigma}_c^{238}(t) \phi(t) + N^{239}(t) \bar{\sigma}_c^{239}(t) \phi(t) \right. \\ & - N^{239}(t) \bar{\sigma}_a^{239}(t) \phi(t) \\ & + N^{241}(t) \bar{\sigma}_c^{241}(t) \phi(t) - N^{241}(t) \bar{\sigma}_a^{241}(t) \phi(t) \\ & \left. - N^{242}(t) \bar{\sigma}_a^{242}(t) \phi(t) \right] \end{aligned} \quad (72)$$

By definition $\bar{\sigma}_a = \bar{\sigma}_c + \bar{\sigma}_f$ or $\bar{\sigma}_c - \bar{\sigma}_a = -\bar{\sigma}_f$. Using this definition the ^{239}Pu and ^{241}Pu terms can be combined resulting in Equation (73).

$$\begin{aligned} \frac{dN^{Pu}(t)}{dt} = & \left[N^{238}(t) \bar{\sigma}_c^{238}(t) \phi(t) - N^{239}(t) \bar{\sigma}_f^{239}(t) \phi(t) \right. \\ & \left. - N^{241}(t) \bar{\sigma}_f^{241}(t) \phi(t) - N^{242}(t) \bar{\sigma}_a^{242}(t) \phi(t) \right] \end{aligned} \quad (73)$$

The differential equation that describes the ^{235}U concentration is:

$$\frac{dN^{235}(t)}{dt} = - N^{235}(t) \bar{\sigma}_a^{235}(t) \phi(t) \quad (74)$$

Note: The creation of ^{235}U by neutron capture in ^{234}U is not represented on the right hand side of Equation (74). The error caused by neglect of this term in the analysis of YANKEE ROWE isotopic concentrations is about 0.5% in the ^{235}U concentration.

We may write a differential equation describing ^{235}d as follows:

$$\frac{d(^{235}\text{d})}{dt} = \frac{d[N^{235}(0) - N^{235}(t)]}{dt} = - \frac{dN^{235}(t)}{dt} \quad (75)$$

Therefore, from Equation (74) and (75) we may write:

$$\frac{d(^{235}\text{d})}{dt} = N^{235}(t) \bar{\sigma}_a^{235}(t) \phi(t) \quad (76)$$

We may divide Equation (73) by Equation (76) providing Equation (76) is not identically zero. The flux cancels out on the right hand side, yielding

$$\frac{\frac{dN(t)}{dt}}{d[^{235}\text{d}(t)]} = \frac{\left[N^{238}(t) \bar{\sigma}_c^{238}(t) - N^{239}(t) \bar{\sigma}_f^{239}(t) - N^{241}(t) \bar{\sigma}_f^{241}(t) - N^{242}(t) \bar{\sigma}_a^{242}(t) \right]}{N^{235}(t) \bar{\sigma}_a^{235}(t)} \quad (77)$$

Equation (77) is an expression for the slope of the total Pu versus ^{235}d curve. The numerator of the fraction on the right hand side of Equation (77) expresses the difference between the production and loss of Pu, in other words the net Pu production. The denominator expresses the loss of ^{235}U . In YANKEE ROWE, at 25,000 MWd/MTU the term $-N^{242}(t) \bar{\sigma}_a^{242}(t)$ is only 1.5% of the Pu terms in the numerator and therefore neglecting this term for the sake of simplicity and clarity is acceptable. Equation (77) then becomes:

$$\frac{dN(t)}{d[{}^{235}\text{d}(t)]} = \left[\frac{N^{238}(t) \bar{\sigma}_c^{238}(t) - N^{239}(t) \bar{\sigma}_f^{239}(t) - N^{241}(t) \bar{\sigma}_f^{241}(t)}{N^{235}(t) \bar{\sigma}_a^{235}(t)} \right] \quad (78)$$

The next step in investigating the reasons for the invariance of Equation (77) with exposure is to somehow relate the denominator in the fraction on the right hand side to the numerator. There must be some fundamental correlation between the numerator and denominator to cause the invariance observed experimentally. The first step in analyzing this correlation is to observe that there is a linear relationship between the fissions in ${}^{235}\text{U}$ and energy in MWd. This linear relationship is graphically displayed in Figure 4 relative to the total fissions. If one considers the hypothetical case where the only fissionable isotope present is ${}^{235}\text{U}$, then.

$$(\text{Total } {}^{235}\text{U fissions}) \left(\frac{\text{Megawatt days}}{\text{fission}} \right) = \text{Total megawatt days} \quad (79)$$

or

$$\left(\int_0^{t_0} N^{235}(t) \bar{\sigma}_f^{235}(t) \phi(t) dt \right) \left(\frac{\text{Megawatt days}}{\text{fission}} \right) = \text{MWd} \quad (80)$$

The total number of fissions in ${}^{235}\text{U}$ is related to the total depletion of ${}^{235}\text{U}$ or ${}^{235}\text{d}$ in the following manner:

$$\begin{aligned} & \left(\int_0^{t_0} N^{235}(t) \bar{\sigma}_a^{235}(t) \phi(t) dt \right) \\ &= \left(\int_0^{t_0} N^{235}(t) [1 + \alpha^{235}(t)] \bar{\sigma}_f^{235}(t) \phi(t) dt \right) \end{aligned} \quad (81)$$

where

$$\alpha^{235}(t) = \frac{\bar{\sigma}_c^{235}(t)}{\bar{\sigma}_f^{235}(t)} .$$

The term $[1 + \alpha(t)]$ varies 2% over the exposure range 0 - 25,000 MWd/MTU for YANKEE ROWE. Thus, the total ^{235}U depletion and total ^{235}U fissions, can be considered to be related in a linear fashion and therefore in this hypothetical situation the total ^{235}U depletion would be linearly related to MWd.

However, the behavior of ^{235}D in YANKEE ROWE is strongly influenced by the presence of Pu as shown in the lower curve of Figure 4. The presence of Pu results in significant nonlinearity with exposure. As the Pu builds up it also fissions and produces power so that at any particular value of total power generated, (MWd), the depletion of U is less than what it would have been in the absence of Pu. In this way, the Pu buildup exerts its influence on the ^{235}U depletion.

Also shown in Figure 4 is a plot of Pu/U versus exposure. The plot of Pu/U versus exposure and ^{235}D versus exposure, are both nonlinear and have very similar curvatures. By dividing the Pu/U by the ^{235}D the nonlinearities tend to cancel each other, with the resulting function Pu/U/ ^{235}D being linear with exposure.

The mathematical expression of the relationship of ^{235}U depletion and Pu buildup is the conservation of power expression:

$$\begin{aligned} \text{Power}(t) = & N^{238}(t) \bar{\sigma}_f^{238}(t) \phi(t) (F^{238}) + N^{235}(t) \bar{\sigma}_f^{235}(t) \phi(t) (F^{235}) \\ & + N^{239}(t) \bar{\sigma}_f^{239}(t) \phi(t) (F^{239}) + N^{241}(t) \bar{\sigma}_f^{241}(t) \phi(t) (F^{241}) \quad (82) \end{aligned}$$

where

F^g = fissions to energy conversion factor for isotope g.

Equation (82) expresses the total power per unit volume in terms of the significant U and Pu fissionable isotopes. Neglecting the other fissionable isotopes causes an error of less than 0.5% in power at 25,000 MWd/MTM for YANKEE ROWE. The fissions to energy conversion factors used for ^{238}U , ^{235}U , ^{239}Pu , and ^{241}Pu , excluding neutrino energy, are those of the Evaluated Nuclear Data File, Version III (ENDF/B-III):

^{238}U : 190.6 MeV/fission

^{235}U : 192.5 MeV/fission

^{239}Pu : 198.6 MeV/fission

^{241}Pu : 200.5 MeV/fission

The ^{239}Pu energy yield is 0.9% less than that of ^{241}Pu and 3.17% more than that of ^{235}U . We may simplify the analysis by assuming that $F^{241} = F^{239}$. This assumption would cause a 0.9% error in the ^{241}Pu power term. We may choose units of power such that $F^{239} = F^{241} = 1$ and $F^{235} = 0.9693$ and $F^{238} = 0.9597$. We may also assume that

Power(t) = constant = P.

A constant power was used in the ZODIAC-G burnup calculations of YANKEE ROWE. The analysis of linearity in the case of a variable power, although more complex, could also be done. With these assumptions Equation (82) becomes:

$$\begin{aligned}
 P = & F^{238} N^{238}(t) \bar{\sigma}_f^{238}(t) \phi(t) + F^{235} N^{235}(t) \bar{\sigma}_f^{235}(t) \phi(t) \\
 & + N^{239}(t) \bar{\sigma}_f^{239}(t) \phi(t) + N^{241}(t) \bar{\sigma}_f^{241}(t) \phi(t) \quad . \quad (83)
 \end{aligned}$$

In order to apply Equation (83), the value of P must be determined. Since YANKEE ROWE is ^{235}U enriched, the contribution of the Pu isotopes to the power at $t = 0$ is zero. The value of P at the beginning of life is

$$P = F^{238} N^{238}(0) \bar{\sigma}_f^{238}(0) \phi(0) + F^{235} N^{235}(0) \bar{\sigma}_f^{235}(0) \phi(0) \quad (84)$$

Solving Equation (83) for $N^{235}(t) \bar{\sigma}_f^{235}(t) \phi(t)$ we have

$$\begin{aligned} N^{235}(t) \bar{\sigma}_f^{235}(t) \phi(t) = & \frac{1}{F^{235}} \left(P - F^{238} N^{238}(t) \bar{\sigma}_f^{238}(t) \phi(t) \right) \\ & + \frac{1}{F^{235}} \left[- N^{239}(t) \bar{\sigma}_f^{239}(t) \phi(t) \right. \\ & \left. - N^{241}(t) \bar{\sigma}_f^{241}(t) \phi(t) \right] \quad (85) \end{aligned}$$

If we multiply Equation (85) by $[1 + \alpha^{235}(t)]$ as was done in Equations (80) and (81) and divide by $\phi(t)$ we have:

$$\begin{aligned} N^{235}(t) \bar{\sigma}_a^{235}(t) = & \frac{[1 + \alpha^{235}(t)]}{F^{235}} \left[\frac{P - F^{238} N^{238}(t) \bar{\sigma}_f^{238}(t) \phi(t)}{\phi(t)} \right. \\ & \left. - N^{239}(t) \bar{\sigma}_f^{239}(t) - N^{241}(t) \bar{\sigma}_f^{241}(t) \right] \quad (86) \end{aligned}$$

Expression (86) relates the denominator term on the right hand side of Equation (77) to the numerator. If we substitute (86) into (78) eliminating $N^{235}(t) \bar{\sigma}_a^{235}(t)$ in (78) we get:

$$\begin{aligned} \frac{Pu}{dN(t)} = & \frac{F^{235}}{1 + \alpha^{235}(t)} \left[\frac{N^{238}(t) \bar{\sigma}_c^{238}(t)}{[P - F^{238} N^{238}(t) \bar{\sigma}_f^{238}(t) \phi(t)]} - \left(\frac{N^{239}(t) \bar{\sigma}_f^{239}(t)}{N^{239}(t) \bar{\sigma}_f^{239}(t)} \right) \right. \\ & \left. + \frac{N^{241}(t) \bar{\sigma}_f^{241}(t)}{N^{241}(t) \bar{\sigma}_f^{241}(t)} \right] \quad (87) \end{aligned}$$

We may make simplifications to (87) in the following manner:

Let:

$$x(t) = N^{239}(t)\bar{\sigma}_f^{239}(t) + N^{241}(t)\bar{\sigma}_f^{241}(t) \quad (88)$$

By adding $-F^{238}N^{238}(t)\bar{\sigma}_f^{238}(t)\phi(t)$ to both sides of Equation (84) we obtain:

$$\begin{aligned} P - F^{238}N^{238}(t)\bar{\sigma}_f^{238}(t)\phi(t) &= F^{235}N^{235}(0)\bar{\sigma}_f^{235}(0)\phi(0) \\ &+ [F^{238}N^{238}(0)\bar{\sigma}_f^{238}(0)\phi(0) \\ &- F^{238}N^{238}(t)\bar{\sigma}_f^{238}(t)\phi(t)] \quad (89) \end{aligned}$$

We may then simplify (89) by neglecting the difference of the ^{238}U terms on the right hand side of (89) which makes an error of less than 1% in YANKEE ROWE data. Expression (89) then becomes:

$$[P - F^{238}N^{238}(t)\bar{\sigma}_f^{238}(t)\phi(t)] = F^{235}N^{235}(0)\bar{\sigma}_f^{235}(0)\phi(0) \quad (90)$$

There is a 2% variation in the value of $F^{235}/1 + \alpha^{235}(t)$ over the exposure range 0 to 25,000 MWd/T in YANKEE ROWE, so one can consider this term as constant to within 2% over that exposure range. Let us assume

$$\frac{F^{235}}{1 + \alpha^{235}(t)} = C \text{ (a constant)} \quad (91)$$

If we rewrite (87) using these simplifications we have the expression we have been seeking for the slope of the $\text{Pu}/^{235}\text{d}$ curve.

$$\frac{\text{Pu}}{d[{}^{235}\text{d}(t)]} = C \left[\frac{N^{238}(t)\bar{\sigma}_c^{238}(t) - x(t)}{N^{235}(0)\bar{\sigma}_f^{235}(0) F^{235}\left[\frac{\phi(0)}{\phi(t)}\right] - x(t)} \right] \quad (92)$$

In Equation (92) we have a functional form which is simple enough for further analysis. A plot of $x(t)$ versus exposure for YANKEE ROWE is shown in Figure 5.

The functional form of the right hand side of Equation (92) can be represented by the form

$$C \left[\frac{a - x}{b - x} \right] \quad (93)$$

Thus since the slope is a function of a , b , and x , the sensitivity of the slope to variations in a , b , and x using partial differentiation is:

$$\frac{\partial f(a,b,x)}{\partial a} = C \left[\frac{1}{b - x} \right] \quad (94)$$

$$\frac{\partial f(a,b,x)}{\partial b} = -C \left[\frac{a - x}{(b - x)^2} \right] \quad (95)$$

$$\frac{\partial f(a,b,x)}{\partial x} = C \left[\frac{a - b}{(b - x)^2} \right] \quad (96)$$

Figure 6 shows the values of a and b as a function of exposure for YANKEE ROWE. For exposures less than 26,000 MWd/MTM, $a < b$ and $b \rightarrow a$ as exposure increases. One would expect $x(t) \leq a$ since a is the source term for the Pu and Pu is still building up. A consequence of this would be $x < b$ since $|b| > |a|$.

It is clear from Equation (94) that as $|x|$ increases and approaches the value of b , the sensitivity of the slope to variations in the parameter a increases.

The behavior of Equation (95) is somewhat more complicated. As $|x|$ increases from zero and approaches the value of a , the sensitivity of the slope to variations in the parameter b approaches zero.

Equation (96) shows that the sensitivity of the slope to the variations in the Pu terms is directly proportional to the difference between a and b . The sensitivity also increases as the Pu terms increase.

The value of the slope at zero exposure is the initial conversion ratio. This can be seen from Equation (78) with $x(t) = 0$.

$$\frac{\frac{Pu}{dN(0)}}{d[{}^{235}d(0)]} = \frac{N^{238}(0)\bar{\sigma}_c^{238}(0)}{N^{235}(0)\bar{\sigma}_d^{235}(0)} \quad (97)$$

Figure 9 shows the variation of $\frac{Pu}{dN(t)/d[{}^{235}d(t)]}$ for YANKEE ROWE as a function of exposure. If this is a linear relationship, the curves in Figure 9 should be horizontal, since a linear curve has a constant slope. It can be seen that the ratio varies about 6% over the exposure range of 0 - 25,000 MWd/MTU. Figure 9 also shows that when ${}^{242}Pu$ is not included in the definition of Pu the variation in the slope of Pu versus ${}^{235}d$ is reduced by 2.4% over the exposure range, 7,500 MWd/MTM to 25,000 MWd/MTM.

It is interesting to compare the variation of $\frac{Pu}{dN(t)/d[{}^{235}d(t)]}$, as expressed in Equation (92), with the variation of $Pu/U/{}^{235}D$, which is the empirical relationship used in MIST. The curves for $\frac{Pu}{dN(t)/d[{}^{235}d(t)]}$ are obtained by inserting values obtained by a ZODIAC-G calculation of Yankee Rowe into Equation (92). The curves for $Pu/U/{}^{235}D$ are obtained by utilizing the isotopic information from the same ZODIAC-G calculation of Yankee Rowe and plotting the ratio of total plutonium/total uranium/ ${}^{235}D$ at various exposures during the depletion calculations. In order to facilitate the comparison, the ratio $Pu/U/{}^{235}D$ is normalized to $\frac{Pu}{dN(t)/d[{}^{235}d(t)]}$ at 7500 MWd/MTU in Figure 9.

The variation of $Pu/U/{}^{235}D$ is about 3% rounded to the nearest percent over the exposure range. When ${}^{242}Pu$ is neglected from $Pu/U/{}^{235}D$,

FIGURE 9. $\frac{dN_{(t)}^{Pu}}{d(^{235}d_{(t)})}$ and $Pu/U/^{235}D$ Versus Exposure
 for Yankee Rowe Cores I and II

the variation is reduced to about 0.5% in the exposure range of 7,500 - 25,000 MWd/MTM. The reduced variation of $\text{Pu/U}/^{235}\text{D}$ is caused in part by the change in total U of about 4% from 0 - 25,000 MWd/MTU, (see Equation 4) and thus the choice of $\text{Pu/U}/^{235}\text{D}$ from practical safeguards considerations has been a judicious choice resulting in a cancellation of much of the variation found in the ratio

$$\frac{\text{Pu}}{dN(t)} \frac{dN(t)}{d[^{235}\text{d}(t)]} \text{ as investigated calculationally.}$$

In addition, the linearity of Pu/U versus ^{235}D has been related directly to Equation (92) which can be used to investigate the sensitivity of $\text{Pu/U}/^{235}\text{D}$ to changes in the different terms of which it is composed. The knowledge of the limits of the linearity of $\text{Pu/U}/^{235}\text{D}$ establishes the degree of accuracy that one may expect from this safeguards tool. For example, at high exposures, >30,000 MWd/MTU, one should expect a slight increase in the slope of the line, (see Figure 9). With the slope increase identified, it is easy to account for it. Also, the initial ^{235}U enrichment and flux spectrum have a significant influence on the linearity property. The initial ^{235}U enrichment in D_2O and H_2O moderated reactors range from the 0.7% enrichment found in natural U to between 4 and 5% in enriched U in H_2O moderated reactors. The variation of the initial ^{235}U enrichment will affect $F^{235}\text{N}^{235}(\text{o})\bar{\sigma}_f^{235}(\text{o})\left[\frac{\phi(\text{o})}{\phi(t)}\right]$ and therefore $\text{Pu/U}/^{235}\text{D}$ will vary inversely with the value of the initial ^{235}U enrichment. There will be some compensating variation in $\bar{\sigma}_f^{235}(\text{o})$ due to flux spectrum changes. This influence of initial enrichment on the slope of the $\text{Pu/U}/^{235}\text{D}$ is shown experimentally in Figure 7. It can be seen from Figure 7 that the various reactors are ranked according to enrichment by the slope. The one exception is the VAK⁽⁸⁾BWR where it appears the effect of the flux spectrum is sufficient to change the ordering of the reactor by enrichment. The effect of decreasing the effective ^{235}U fission cross section through increasing the void fraction is shown in Figure 8. Increasing the void fraction hardens the flux spectrum through a decrease in moderator to fuel ratio and produces the

increase in slope. In both cases the curves conform to the functional dependence predicted by Equation (92). Other parameters which affect the flux spectrum, such as fuel dimensions and type of cladding, will also affect the ratio. However, their affect can also be investigated through Equation (92).