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SPECTRUM AVERAGED CROSS SECTIONS DEDUCED FROM
BURNUP DATA AND THEIR APPLICATION TO METHODS VERIFICATION (a)

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ABSTRACT

The determination of ratios of spectrum averaged cross sections from isotopic concentrations is described along with their application to verification of analytical reactor design tools. The determination is made using specially developed least squares computer programs which fit the mathematical transmutation relationships to isotopic concentrations measured in irradiation experiments. These data are utilized as a basis for evaluating the accuracy of calculational methods used to predict burnup behavior of nuclear fuels. Data obtained from irradiation experiments using plutonium-aluminum alloy fuel are given to illustrate the techniques and demonstrate how these data are used to verify calculational methods.

INTRODUCTION

The accurate prediction of fuel burnup is important for producing reliable and low cost power with nuclear reactor power systems. The accurate calculation of fuel burnup is complicated by the number of variables which are present in the mathematical representation of the processes which affect the transmutations occurring in the fuel. The success of the burnup calculation depends on the validity of the theoretical and mathematical models along with the nuclear data used as input in the calculation. The accuracy of the burnup prediction can be verified through correlations of experimental burnup data and results from burnup calculations.

The usual approach to solving the burnup problem is to acquire data on the variation of fuel isotopic composition as a function of irradiation time and use these data to evaluate the merit of burnup calculations. In terms of the theoretical and mathematical transmutation relationships which describe these data, the time integral of the flux and the time dependent spectrum average nuclear cross sections are problem variables. An approach was developed at Battelle-Northwest to build upon this data base of time dependent isotopic compositions by eliminating one of these variables from the problem. The approach was to eliminate the flux-time variable from direct consideration.

The transmutation equations are cast as ratios to eliminate the flux-time variable and these equations fitted to the measured isotopic concentrations to yield ratios of spectrum averaged neutron cross sections (hereafter referred to simply as cross section ratios). The

fitting is accomplished using specially developed least squares fitting techniques.

The intent was to generate data which focused on evaluation of the theoretical methods and/or the neutron cross sections used in burnup calculations. More specifically, the principal objective was to obtain data which can be used to verify the accuracy of the starting point calculation of neutronic reactor analysis, namely, the multigroup neutron spectrum in a unit cell of the reactor. Thus, the isotopic data from which the cross section ratios are deduced have to be obtained from fuels resulting in those regions of the reactor which are typical of the unit cell of interest. Thus, a basic assumption is that the data used in the analysis come from samples irradiated in the same nuclear environment. The technique is not applicable for samples from two locations of the reactor where neutron spectra are different for a given irradiation such as near control rods or leakage boundaries and in the center of fuel bundles.

The burnup data to test this technique have been obtained by chemical assays and mass spectrometric measurements of aluminum-plutonium (Al-Pu), uranium oxide (UO_2), and mixed uranium-plutonium oxide ($\text{UO}_2\text{-PuO}_2$) fuel assemblies irradiated in D_2O and H_2O moderated power reactors. These measurement techniques are described in Section II.

The lack of a statistical analysis method with sufficient versatility to analyze the data made it necessary to first solve the statistical problem as described in Section III. The data analysis method is described in Section IV and an application for Al-Pu alloy fuels which were irradiated in the Plutonium Recycle Test Reactor (PRTR) is also given.

The utilization of these data in verification of burnup calculations is described in Section V where the results of the correlation for the Al-Pu alloy fuel are given in Table IV.

TRANSMUTATION EQUATIONS

The equations used in deducing cross section ratios from burnup data are the differential equations,

$$\frac{dN^i}{dt} = -\lambda^i N^i - \hat{\sigma}_a^i N^i \phi + \hat{\sigma}_c^i N^{i-1} \phi \quad (1)$$

These equations equate the concentration change of an isotope with time to the loss by decay, the loss by absorption, and the gain by capture. Other sources of an isotope such as external additions or from fission are excluded from the equations so that only unreplenished fuel materials are being considered. Also, any contribution from the decay of a parent isotope is negligible for the isotopes considered and has been ignored. The isotope concentrations N^i which are involved are N^{25} , N^{26} , and N^{28} for the uranium isotopes of mass 235, 236, and 238, and N^{49} , N^{40} , N^{41} , and N^{42} for the plutonium isotopes of mass 239, 240, 241 and 242. The cross section values are flux and volume averaged values and are defined as

$$\hat{\sigma}_a^i = \frac{\int_{E=0}^{\infty} \int_V \sigma_a^i(E) \phi(\underline{r}, E) dE d\underline{r}}{\int_{E=0}^{\infty} \int_V \phi(\underline{r}, E) dE d\underline{r}} \quad (2)$$

Division of all equations (1) by

$$\frac{dN^j}{dt} = -\hat{\sigma}_a^j N^j \phi \quad (3)$$

eliminates the flux ϕ and time t variables except in the small terms which describe the decay. The result is another set of differential equations involving cross section and concentration ratios,

$$\frac{dN^i}{dN^j} = + \frac{\lambda^i N^i}{\hat{\sigma}_a^j N^j \phi} + \frac{\hat{\sigma}_a^i N^i}{\hat{\sigma}_a^j N^j} - \frac{\hat{\sigma}_c^{i-1} N^{i-1}}{\hat{\sigma}_a^j N^j} \quad (4)$$

Equation (3) will describe the ^{235}U changes in the case of a UO_2 or $\text{UO}_2\text{-PuO}_2$ system and ^{239}Pu changes for a Pu system. In this way the ^{235}U and ^{239}Pu concentrations are chosen as burnup indicators. Their concentration changes are the only ones which are described as exponential functions of the exposure ϕt .

Two separate methods, each making extensive use of specially developed least squares fitting programs have been developed to obtain ratios of flux and volume integrated cross sections from equation (4).

In one method (differential), the several burnup equations described by equations (4) are solved by separation of the variables and integration over a 10^{-5} at/b-cm change in N^j . Equations of the form

$$\begin{aligned} \Delta N^i = & - \frac{\hat{\sigma}_c^{i-1}}{\hat{\sigma}_a^j} \int \frac{N^{i-1}}{N^j} dN^j \\ & + \frac{\hat{\sigma}_a^i}{\hat{\sigma}_a^j} \int \frac{N^i}{N^j} dN^j + \frac{\lambda^i}{\hat{\sigma}_a^j} \int \frac{N^i}{\phi N^j} dN^j \end{aligned} \quad (5)$$

are obtained where the $\hat{\sigma}^j$ and $\hat{\sigma}^i$ are considered to be constant over the small region of integration. Equations (5) are then successively solved for the cross section ratios which best fit the experimental data in the least squares sense.

Regression equations of inverse polynomial form

$$(N^i/N^j) = \sum_{k=0}^n A_k (N^j \times 10^6)^{-k} = f^i(N^j) \quad (6)$$

are used to represent the data. The values of A_k which yield the best fit to the data together with their uncertainties are used to determine cross section ratios by integration over an appropriate burnup increment. Since

$$N^i = N^j f^i(N^j) \quad (7)$$

then

$$\Delta N^i = N_b^j f^i(N_b^j) - N_a^j f^i(N_a^j) \quad (8)$$

where a and b are the integration limits of the burnup increment. The cross section ratios are obtained by solving equations (5) using equations (6), (7), and (8).

In another method (integral), analytical solutions can be obtained for equations (4) by proper choices of integrating factors if the cross sections $\hat{\sigma}^i$ relative to $\hat{\sigma}^j$ remain constant as a function of exposure. In the cases studied this is approximately true for all cross sections except $\hat{\sigma}_a^{40}$. An acceptable empirical form has been found for the ^{240}Pu ratio which still allows analytical solutions to be obtained for equations (4). This extension makes it possible to obtain a set of integrated equations which are amenable to being fit to experimental data by the method of least squares.

A least squares fit of the analytical solutions to the experimental data is considerably more complex than is ordinarily encountered. First, only variables N^i are considered as opposed to a dependent variable and one or more independent variables. The uncertainties associated with each data point must be described by a matrix as opposed to the simpler situation where only the error in the dependent variable is required. Lastly, a series of related equations must be fit simultaneously to a set of experimental data. Before this method could be used, it was necessary to develop a least squares fitting routine with the required sophistication to solve these problems.

Each method has advantages and disadvantages to the extent that the two methods complement each other. Initially, the experimental data was analyzed in each way. The first method using differential equations has the advantage of yielding cross section ratios as a function of exposure. The requirements of assuming a value for one of the ratios and of neglecting correlated uncertainties between various N^i for a given sample are disadvantages. On the other hand, the second method using integrated equations simultaneously fits all the data and correctly accounts for correlated uncertainties. The integrated equation approach also provides a rigorous statistical analysis of the data and required no a priori values to be assumed for the cross section ratios. The disadvantage of the method lies in the restrictive form of the cross section ratios which are necessary to obtain analytical solutions. However, in most cases empirical forms can be found which adequately meet the requirements of the analysis.

A sensitive assessment of the measurement accuracy is provided by the extent of the agreement of the results of the two methods compared with the statistical uncertainty. In addition, the total fissions can be calculated after the cross-section ratios are obtained by either method. Comparison of calculated values of the total fissions or percent depletion with those obtained experimentally by ^{137}Cs or other analysis provides further assessment of measurement accuracy.

TRANSMUTATION MEASUREMENTS

Fuel composition and concentrations have been measured as a function of exposure and used in correlations with analytical results. The measurements include data from 19-rod clusters irradiated in the PRTR and individual rods irradiated in the Experimental Boiling Water Reactor (EBWR).^(1,2) The correlations include data from the Saxton Reactor⁽³⁾ and Yankee Reactor⁽⁴⁾ in addition to that from PRTR and the EBWR.

The types of fuels that are being used and their exposures are listed in Table I. All exposures are given in MWd/MTU except for the Al-Pu systems for which exposures are given as atom percent depletion of the Pu.

The quantities determined by destructive analysis of the fuels are sample weights and dissolution volumes, ^{137}Cs activity, U, Pu, and ^{148}Nd contents, and U and Pu isotopic compositions.

TABLE I

FUEL TYPES AND EXPOSURES

PRTR (D₂O)

<u>Al-Pu</u> ⁽⁵⁻⁷⁾	<u>Initial</u> <u>²⁴⁰Pu (wt%)</u>	<u>Maximum</u> <u>Exposure</u> ^(a)
1.8 wt% Pu	6	50
2.6 wt% Pu	16	52
<u>UO₂ (Natural)</u> ^(8,9)		15000
<u>UO₂-PuO₂</u> ^(10,11)		
0.48 wt% PuO ₂	17	11700
1.0 wt% PuO ₂	8	5800
2.0 wt% PuO ₂	8	9700

EBWR (BWR)

<u>Al-Pu</u> ^(12,13)		
3.4 wt% Pu	8 and 26	25
<u>UO₂ (6% Enriched)</u> ^(12,13)		4800
<u>UO₂ (Depleted)-PuO₂</u> ^(12,13)		
1.5 wt% PuO ₂	8, 20 and 26	6800

Yankee Reactor (PWR)

<u>UO₂</u> ^(14,15)	3.4 wt% ²³⁵ U	32000
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Saxton (PWR)-Core II

<u>UO₂ (Natural)-PuO₂</u> ^(16,17)		
6.6 wt% PuO ₂		29000

(a) All exposures are in MWd/MTU except for the Al-Pu systems for which exposures are in atom percent depletion of the PU.

Radiometallurgy Procedures

After approximately two months cooling, an entire cluster or rod is shipped to the Radiometallurgy Laboratory for cutting and dissolution. After a rod is sawed into segments, two samples of fuel and cladding each approximately 1/2 in. long are cut by an abrasive wheel from a segment and weighed on an analytical balance.

For Al-Pu fuels, one sample is added to 150 ml solution consisting of 1 M HNO_3 , 0.015 M $\text{Hg}(\text{NO}_3)_2$, and 8×10^{-6} M CsNO_3 and heated to 60°C. The Cs^+ ions minimize the exchange of ^{137}Cs for materials in the glassware. The Hg^{2+} ions act as a catalyst. Heating is continued until a vigorous reaction starts. Then 85 ml of a solution containing 15.6 M HNO_3 and heated to 60°C is added. The solution removes the core material leaving the Zircaloy cladding practically unattacked. The weight of the clean cladding is obtained and used to determine the fuel weight. A standard cladding sample weighing 5.886 gm was found to lose 40 mg after being submitted to the dissolution process. After removing the cladding, the solution of fuel is diluted to 250 ml with 2M HNO_3 . The volume, and temperature are measured and the density (gm fuel/ml solution) of the fuel in solution is calculated. The other sample is stored for later analysis if needed.

For uranium oxide or mixed oxide fuels one sample is dissolved in 117 ml of solution containing 13.5 M HNO_3 , 8×10^{-6} M CsNO_3 and 0.023 M HF. In order to keep the HF from attacking the glass containers, AlNO_3 is added. The solution is heated but kept below boiling until there is no more reaction taking place. After dissolution of the fuel material the

cladding is removed and weighted. Another 10 ml of water containing 1-2 drops of HF is added to the fuel solution if needed for complete dissolution. The solution is reheated for ~ 2 hours to assure that all the fuel is dissolved. After cooling, enough 2 M HNO_3 is added to bring the total to 250 ml. The volume and temperature are measured and the density of the fuel in solution is calculated.

Analytical Chemistry Procedures

Samples of unirradiated fuel and ~ 15 ml solutions of the irradiated fuel are sent to the Analytical Laboratory for analyses. The ^{137}Cs content is measured by gamma-ray spectrometry. The fission product ^{148}Nd content is determined by the isotopic dilution-mass spectrometry technique.⁽¹⁸⁾ The plutonium and uranium contents and isotopic abundances are determined by isotopic dilution and mass spectrometry. The plutonium is also measured by alpha counting, and the plutonium and uranium are measured by controlled-potential coulometric titration.^(19,20) The isotopic dilution-mass spectrometry method for plutonium and uranium provides the best accuracy. The results from the coulometric titration procedure for plutonium and uranium, and the alpha counting procedure for plutonium provide independent verification of results obtained by isotopic-dilution mass spectrometry.

Coulometric titration of plutonium is accomplished using a platinum electrode cell with an electrolyte of 1M HCl containing 10 grams per liter Al^{+3} and a small quantity of urea (50 μl of saturated urea solution is added to 5 ml of electrolyte). The Al^{+3} eliminates

interference from fluoride ions and the urea eliminates interference from nitrite ions. The plutonium sample size may range from 100 micrograms up to several milligrams. The best accuracy is obtained with the larger samples. The plutonium and iron in the sample are reduced to Pu^{+3} and Fe^{+2} , respectively, at a potential of + 0.3 volts relative to a saturated calomel electrode. The potential is raised to + 0.84 volts to oxidize the plutonium and iron to Pu^{+4} and Fe^{+3} . The plutonium is then quantitatively reduced to Pu^{+3} at a potential of + 0.56 volts. The plutonium content is determined from the total change obtained by integrating the reduction electrolysis current and subtracting background currents during the last reduction step. Also, a correction is made for the very small fraction of the Fe^{+3} reduced during the reduction at + 0.56 volts. In the absence of contaminants which affect the Pu titration, accuracies ⁽²¹⁾ of ~ 2% are obtained from solutions having greater than 50 mg/l Pu.

Uranium is determined coulometrically using a mercury electrode cell and a 1M H_2SO_4 electrolyte. The normal sample size taken for uranium is 2 to 5 mg. The uranium is oxidized to U^{+6} by adding 0.1N ceric sulfate solution. The sample is pre-reduced at a potential of + 0.05 volts relative to a saturated calomel electrode until a residual current is reached. This step removes constituents that reduce at a more positive potential than uranium. The potential is decreased to - 0.30 volts and the uranium quantitatively reduced from U^{+6} to U^{+4} . The uranium content is determined from the total charge obtained by integrating the reduction electrolysis current and subtracting background currents during the last reduction step.

The results of the coulometric titration procedures provide independent verifications of the Pu and U contents obtained from the isotopic dilution mass spectrometry method. The verification is made for 10% of the oxide samples and 25% of the Al-Pu samples. The plutonium from a measured portion of the sample is removed by thenoyltrifluoroacetone⁽²²⁾ (TTA) extraction. The amount of plutonium is determined to an accuracy of $\pm 1\%$ by counting α particles emitted by the plutonium. The total alpha-particle activity of the plutonium is measured with 2π alpha-Simpson proportional counter calibrated with a known Pu standard. Energy analysis of the emitted particles using a surface barrier silicon diode detector (or formerly a Frisch grid ionization chamber) coupled to a multichannel analyzer allows subtraction of the α activity⁽²³⁾ of ^{238}Pu from the total activity. The weight of the Pu, excluding ^{238}Pu , is calculated from the value for the remaining activity using a weighted average specific α activity based on the isotopic abundances for ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu determined by mass spectrometry.

The cesium is removed from a measured portion of the sample by tetraphenylborate (TPB) extraction.⁽²⁴⁾ The 622 KeV gamma-ray activity emitted from the cesium (^{137}mBa daughter) is counted in a NaI well crystal. Corrections are made for ^{134}Cs by energy analysis and the result is normalized to a SRM-4233, National Bureau of Standards, cesium standard to obtain the ^{137}Cs atoms/ml of fuel solution.

A correction for ^{137}Cs decay is made because the analyses are

completed months after the irradiation was terminated. The cesium activity (^{137}Cs) is converted to ^{235}U and Pu fissions using fission yields ^(25,26) of 0.0622 ± 0.0014 , 0.0648 ± 0.0019 , and 0.0662 ± 0.0033 for ^{235}U , ^{239}Pu , and ^{241}Pu , respectively and a half-life ⁽²⁷⁾ of 29.68 ± 0.10 years ^(a) for ^{137}Cs . The ^{148}Nd content is also used to determine fissions especially when possibilities of cesium migration are present. In this case, fission yields ^(25,26) of 0.0169 ± 0.0003 , 0.0165 ± 0.0005 , and 0.0188 ± 0.0009 are used for ^{235}U , ^{239}Pu , and ^{241}Pu , respectively.

Mass Spectrometry Procedures

Determinations of the U and Pu concentrations and isotopic compositions are made ⁽²⁸⁾ using two mass spectrometers with 1st order angular focusing. Each mass spectrometer has a 12-in. radius, 60 degree magnet sector and uses a surface ionization source. A single filament of carbonized rhenium or a triple filament of rhenium metal produces metal ions from samples adjusted to contain 10-50 ng of material.

The same instruments are used for the determination of fission product ^{148}Nd . A single pre-carbonized rhenium filament or a single rhenium metal filament is loaded with about 70 ng of neodymium. In the case of the metal filament, 10-20 microliters of 0.005 M sucrose solution is dried on the filament with the sample. Metal ions are produced from both types of filaments.

The ion beam of constant energy of 12 kV is swept past the detector ten times in each direction to obtain the results. The detector is an electron multiplier operated as an integrated current device. The gain is mass and relative intensity dependent so the instrument is calibrated

with isotopic standards of uranium and plutonium from the National Bureau of Standards, and with a natural neodymium standard for neodymium.

Samples of U and Pu are prepared for mass spectrometry by separation on a single Dowex-1 X 4, 200-400 mesh anion-exchange column using a sequential elution technique. (In an earlier procedure the Pu was separated and purified using a combination of tri-iso-octylamine and thenoyltrifluoroacetone extractions.)

Before the separation process and after being diluted a sample of fuel solution from Radiometallurgy is spiked with a solution containing known amounts of ^{233}U and ^{242}Pu . A second sample is processed but without the spike solution. The spike solution contains a known amount of ^{242}Pu which has been calibrated with a metallic ^{239}Pu standard from the National Bureau of Standards. It also contains a known amount of ^{233}U calibrated with a natural uranium standard from the Bureau of Standards. The separated solutions of U, (U plus ^{233}U) spike, Pu, and (Pu plus ^{242}Pu) spike are then analyzed separately for their isotopic compositions. The uranium and plutonium concentrations in the sample are calculated from the known spike volume and concentrations, the known spiked sample volume, the measured isotopic compositions, and the measured ^{238}U to ^{233}U and ^{239}Pu to ^{242}Pu atom ratios in sample, spiked sample, and spike solution.

A Nd fraction is separated for mass spectrometry using the anion-exchange procedure of Rider, et al.⁽¹⁸⁾ The same sample and spiked sample used for the U and Pu determinations is used for the Nd. In this case

the spike solution contains a known amount of ^{150}Nd in addition to the known amounts of ^{233}U and ^{242}Pu . The fission product ^{148}Nd concentration in the sample is calculated from the known spike volume and concentration, the known spiked sample volume, the known ^{150}Nd to ^{142}Nd and ^{148}Nd to ^{142}Nd atom ratios in natural Nd, and the ^{142}Nd to ^{150}Nd and ^{148}Nd to ^{150}Nd atom ratios measured in the sample, spiked sample, and spike solution. The ^{142}Nd , a shielded nuclide which is not produced directly in fission, is used to correct for contamination from natural Nd. ⁽¹⁸⁾

It is of interest to note that with the use of a "triple spike", that is a single spike solution containing known amounts of ^{233}U , ^{242}Pu and ^{150}Nd , that the ratios of Pu to U and ^{148}Nd to U obtained by the isotopic dilution-mass spectrometry method are independent of the spike and sample volumes used.

Data Compilation

Because of the large volume of data collected it is convenient to compile the data for each sample, perform subsequent calculations and plot the results using computers. The Burnup Data Analysis Code (ABC) ⁽⁶⁾ and the HADES code ⁽²⁹⁾ or Isotopic Dilution Code (ISODIL) ⁽³⁰⁾ are used to compile the data and determine the burnup parameters and their one standard deviation uncertainties. The data which have been compiled are:

For Al-Pu Samples

Descriptive and historical data

Sample weight, cladding weight, dissolution volume

^{137}Cs activity, alpha particle activity

%²³⁸Pu alpha activity

g/l Pu by coulometric titration

Atomic ratios of isotopes.

For UO₂ and UO₂-PuO₂ Samples

Same as for Al-Pu except the alpha particle and %²³⁸Pu alpha activity are not compiled when atom ratios determined from uranium and plutonium samples are used.

The parameters determined from the data are:

For Al-Pu Samples

Atom ratios and atom percents of Pu isotopes (²⁴¹Pu corrected for decay)

Pu atoms left (from α counts or coulometer results) and Pu atoms depleted (from ¹³⁷Cs activity) per milliliter

Atoms ¹³⁷Cs per milliliter corrected for decay

% depletion (from Pu atoms depleted and left)

Pu atom densities (from percent depletion and initial atom fractions Pu)

Comparison of sample concentration (weight and volume data vs Pu atoms left and depleted)

Ratio ¹³⁷Cs to Pu α particle activity

and

For UO₂ and UO₂-PuO₂ Samples

Atom ratios and atom percents of U and Pu Isotopes (²⁴¹Pu corrected for decay).

Pu/U ratio from Pu α counting and atom ratios and fuel weight and composition of the sample; or from isotopic dilution technique where Pu/U ratio is determined from atom ratios of the spiked U and Pu solutions.

Beginning of life (BOL), end of life (EOL), and net production of U and Pu isotopes in gms per gm (or metric ton) of beginning of life uranium (determined from BOL and EOL atom ratios and Pu/U ratios).

Burnup = gms of material fissioned for ²³⁵U, ²³⁸U (fast), ²³⁹Pu and ²⁴¹Pu per gm (or metric ton) of beginning of life U.

The burnups are also converted into MWd/MTU of U and totaled.

Fissions are determined from ^{137}Cs (corrected for decay) and $^{235}\text{U}_{\text{Fis}} =$
 $-\Delta\text{U}^{235} - \Delta\text{U}^{236}$ and $\text{Pu}_{\text{Fis}}^{241} = \Delta\text{Pu}^{242}/\alpha^{41}$

Plots of the

$^{241}\text{Pu}/^{239}\text{Pu}$ and $^{242}\text{Pu}/^{239}\text{Pu}$ atom ratio vs. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio

^{137}Cs to alpha activity ratio vs. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios

Percent depletion vs. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio

$^{236}\text{U}/^{238}\text{U}$ atom ratio vs. $^{235}\text{U}/^{238}\text{U}$ atom ratio

$^{240}\text{Pu}/^{239}\text{Pu}$, $^{241}\text{Pu}/^{239}\text{Pu}$ and $^{242}\text{Pu}/^{239}\text{Pu}$ atom ratio vs.

$^{235}\text{U}/^{238}\text{U}$ and $^{236}\text{U}/^{238}\text{U}$ atom ratios.

$^{241}\text{Pu}/^{239}\text{Pu}$ and $^{242}\text{Pu}/^{239}\text{Pu}$ atom ratios vs. $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratio

^{137}Cs to alpha activity ratio vs. $^{235}\text{U}/^{238}\text{U}$ and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios

Percent depletion vs. $^{236}\text{U}/^{238}\text{U}$ and $^{240}\text{Pu}/^{239}\text{Pu}$ atom ratios

Atom density of each isotope vs. percent depletion

are included as output. Uncertainties corresponding to one standard

deviation are determined from error equations derived from normal

procedures for propagation of errors. ⁽³¹⁾ It is assumed that no correlation

of errors of the Pu or U atom ratios exist. However, uncertainties due

to mass spectrometer bias, sample handling, separation procedures and

^{241}Pu decay are included.

Analytical functions are fit to the data by the method of least

squares as a method to monitor the data and to obtain more accurate

averaged data. For this purpose the data is represented as a function of

vertical position in the reactor. The measurement quantities ^{137}Cs

(atoms/ml), fuel density (gm/ml), and α activity (d/min-ml) result in ^{137}Cs

(at/gm), α activity (d/min-gm) and the $^{137}\text{Cs}/\alpha$ ratio (at-min/d). The

measurements and data which deviate from the fitted line may do so because

of either inaccurate ^{137}Cs or α particle determination. If the ^{137}Cs

or α activity is high the ratio would be high or low respectively. If

a low fuel density is present the ^{137}Cs (at/gm) and α activity (at/gm)

will both be higher than the fitted curve. Thus samples can be chosen for reanalysis or specific measurement techniques can be identified which need improving.

STATISTICAL ANALYSIS OF TRANSMUTATION MEASUREMENTS

One of the approaches used in analyzing the data from the transmutation measurements is to eliminate the imprecisely known exposure (ϕt) from direct consideration and to determine parameters for equations which describe the data. This is accomplished by casting analytical solutions

$$g(\underline{y}, \underline{p}) = 0 \quad (9)$$

to equations (4) in terms of the interrelated isotopic concentrations. These solutions, as implicit function of exposure, consist of any non-redundant set of $J-1$ equations obtained by algebraic elimination of the exposure t from the J -fold set of isotopic concentrations $\underline{y}$. The isotope concentration vectors $\underline{y}$ as explicit functions of exposure are of matrix exponential form. (32)

$$\underline{y}(t) = \underline{y}(0) \exp(tH) + Q [\exp(tH) - 1]/H.$$

The theory parameters $\underline{p}$ (which are related to the cross section ratios) comprise initial concentrations $\underline{y}(0)$, source sink vector $\underline{Q}$, and transmutation rate matrix H . A least squares analysis of the experimental data endeavors to evaluate the parameters $\underline{p}$ (which are related to the cross section ratios) by optimal minimization of the residual differences (inverse variance weighted) between the measured data and the theory equations (9). The statistical analysis problem had to be solved before this approach could be used with the data because of the apparent absence in the literature of any method having sufficient versatility for the problem. The solution of such a complex statistical problem is a result of the burnup program and has an effect extending to most other general statistical problems.

The foundation of the analysis logic is the minimization of the mean square misfit quantity

$$Q(\underline{p}) = \sum_{m=1}^M \underline{q}(\underline{y}_m, \underline{p}) \cdot \left[\sum_{i=1}^J \sum_{j=1}^J \{ \partial \underline{q}(\underline{y}_m, \underline{p}) / \partial y_m^i \} U_m^{ij} \{ \partial \underline{q}(\underline{y}_m, \underline{p}) / \partial y_m^j \} \right]^{-1} \underline{q}(\underline{y}_m, \underline{p}). \quad (10)$$

by imposing the conditions

$$\partial Q / \partial \underline{p} = 0 \text{ and } \partial^2 Q / \partial \underline{p} \partial \underline{p} = \text{positive definite}. \quad (11)$$

The following subsections describe the development of the least squares misfit quantity, construction of the nonlinear theory equations, and the assignment of statistical uncertainties. All are required for the statistical analysis of this complex problem.

Mean Square Misfit

A three dimensional projection of a hypothetical four dimensional burnup study of ^{239}Pu is shown in Fig. 1 and provides a simple introduction to the complexity of the problem. Each of the axes correspond to one of the isotopic concentrations which describe a data point: either ^{239}Pu , ^{240}Pu or ^{241}Pu . The fourth axis $y_4 = N(\text{Pu-242})$ is not shown but is present both in measurement and theory. The data from a single burnup sample defines a measurement vector, $\underline{y}$ and is shown surrounded by its measurement error ellipsoid. The ellipsoid is analytically described by a measurement variance-covariance matrix

$$U = \langle (\delta \underline{y})(\delta \underline{y})^T \rangle. \quad (12)$$

The U can be contrasted with the case for the more familiar least squares problem which has an error assigned to only one of the variables. The ellipsoid of the complex problem represents the one standard deviation error region in the same sense as does the error flag for the simple case.

Figure 1. A Three Dimensional Projection of the Burnup of ^{239}Pu .

The spiral line represents the least squares fit to the data under the assumption of an appropriate analytical description of the burnup process. The tube surrounding the spiral represents the statistical uncertainty in the fitted spiral as determined from the fitting process.

The construction of equation (10) can be verified by an inductive line of thought. First, when only one variable has an error associated with it, the mean square misfit $Q(\underline{p})$ is the sum of the residuals divided by their variance.

$$Q(\underline{p}) = \sum_{m=1}^M \left[y_m - y_T(\underline{p}) \right]^2 / \sigma_m^2 = \sum_{m=1}^M dy_m U_m^{-1} dy_m \quad (13)$$

where m and T indicate measurement and theory quantities respectively.

In the more complex case dy is a vector and U is a matrix rather than a number so that equation (13) is composed of terms of the form

$$(\underline{dy}) \cdot U_m^{-1} \cdot (\underline{dy}) = \sum_{i=1}^J \sum_{j=1}^J (dy^i) U_{ij}^{-1} (dy^j). \quad (14)$$

Equation (14) reduces to the terms of equation (13) for the more familiar least squares problem in which $J = 1$. In matrix form, U includes both information about the errors on each of the variables (diagonal terms) and information about error correlations between variables (off-diagonal terms). Transformed from the measurement coordinates $\underline{y}$ to the misfit coordinates $\underline{q}$, this expression becomes

$$(\underline{dq}) \cdot \left[\sum_{i=1}^J \sum_{j=1}^J (\partial \underline{q} / \partial y^i) U^{ij} (\partial \underline{q} / \partial y^j) \right]^{-1} \cdot (\underline{dq}) . \quad (15)$$

Since the theory equations have been cast in the implicit form $q(\underline{y}, \underline{p}) = 0$ with one less q variable than y variable, the misfit differentials

$$d\underline{q} = \sum_{j=1}^J \left[\partial q(\underline{y}, \underline{p}) / \partial y^j \right] dy^j$$

span only the $(J-1)$ dimensional subspace normal to the theory curve. Consequently, equation (14) transforms to a degenerate form which ignores the coordinate tangential to the fitted curve. This is exactly what is required for the burnup analysis; transformation of an imprecisely known exposure (ϕt) to an ignorable variable. Replacement of $d\underline{q}$ by $\underline{q}(\underline{y}_m, \underline{p})$ and summing the mean square misfits of equation (15) over all measurements m , completes the construction of the least square misfit, $Q(\underline{p})$ of equation (10).

Maximum Likelihood Nonlinear Theory

The complexity of the nonlinear theory equations (9) is limited only to the existence of the minimal misfit conditions of equations (11) and the misfit quantity equation (10) over the domain of application. For example, they may be

- implicit or explicit functions of the variables $\underline{y}$
- nonlinear in any or all variables $\underline{y}$
- nonlinear in the parameters $\underline{p}$
- degenerate (fewer field equations $q = 0$ than field variables $\underline{y}$).

The search for the minimal misfit $Q(\underline{p})$ proceeds via an iterative

process. The nonlinear differential equations $\partial Q/\partial \underline{p}$ are expanded in truncated Taylor's series

$$\partial Q(\underline{p})/\partial \underline{p} + (d\underline{p}) \cdot (\partial^2 Q(\underline{p})/\partial \underline{p} \partial \underline{p}) = 0$$

and solved for the parameter differences $d\underline{p}$ by matrix inversion

$$d\underline{p} = - (\partial Q(\underline{p})/\partial \underline{p}) \cdot (\partial^2 Q(\underline{p})/\partial \underline{p} \partial \underline{p})^{-1}.$$

These quantities are then used in the algorithm

$$\underline{p} = \underline{p} - (\partial Q(\underline{p})/\partial \underline{p}) \cdot (\partial^2 Q(\underline{p})/\partial \underline{p} \partial \underline{p})^{-1} \quad (16)$$

which, optimistically, provides parameters for an improved least squares fit (i.e., one in which there is a reduction in the misfit value $Q(\underline{p})$).

After each such parameter change, the mean square misfit Q is recomputed from equation (10) and compared to its previous least value. A lower mean square misfit value, indicative of a convergent trend, closes the iterative loop by a recursive traverse of the refinement algorithm of equation (16). A higher mean square misfit value, indicative of a divergent trend, branches the logic first to cubic-expansion of $Q(\underline{p})$ in terms of the last parameter change, and finally to a reversing-and-halving of the last parameter change, in order to provide a two-stage recovery effort. The occurrence of any spill, division by zero, or non-invertible matrix aborts the calculation. Complete agreement of the mean square misfit with its previous least value terminates the iterative process. Output includes the normal principal blades of the misfit second gradient $\partial^2 Q/\partial \underline{p} \partial \underline{p}$ to provide decisive assessment of the positive-definite requirement.

Maximum Likelihood Nonlinear Error Analysis

The third major phase of the statistical development involves the assignment of errors to the parameters and other related quantities. This has been accomplished by a generalization of Fisher's maximum likelihood statistics to the non-linear least squares problem⁽³³⁾. The principal result of the generalization is embodied in the parameter variance-covariance matrix U . The matrix U is twice the matrix inverse of the mean square misfit second gradient, $\partial^2 Q(\underline{p})/\partial \underline{p} \partial \underline{p}$, times the measurement variance expected if one begins over with a new experiment. The latter quantity is the mean square misfit $Q(\underline{p})$ divided by the number $(L \cdot M - K)$ of degrees of freedom for the problem so

$$U = \langle (\delta \underline{p})(\delta \underline{p}) \rangle = 2(\partial^2 Q(\underline{p})/\partial \underline{p} \partial \underline{p})^{-1} Q(\underline{p})/(L \cdot M - K). \quad (17)$$

The L , M , and K are the total number of equations q , data points, and parameters $\underline{p}$ respectively. The root-mean-square uncertainty in each theory parameter can be computed from the diagonal elements of this matrix as

$$\delta p^k = \pm \langle (\delta p^k)(\delta p^k) \rangle^{1/2}. \quad (18)$$

Programs LIKELY and DBUFIT

Thus a set of vector measurements $\underline{y}_m \pm U_m^{1/2}$ are fitted by a set of theory equations $\underline{q}$ and a set of parameters $\underline{p}$ to using a least squares fitting method and allowing a set of parameters $\underline{p}$ to vary until a best fit is obtained. This is accomplished using Program LIKELY and a theory subroutine. The theory subroutine provides the function $\underline{a}(\underline{y}, \underline{p})$ and $\partial \underline{q}/\partial \underline{y}$, and their first and second order parametric gradients $\partial \underline{q}/\partial \underline{p}$, $\partial^2 \underline{q}/\partial \underline{y} \partial \underline{p}$, $\partial^2 \underline{q}/\partial \underline{p} \partial \underline{p}$, and $\partial^3 \underline{q}/\partial \underline{y} \partial \underline{p} \partial \underline{p}$ which are needed for the fitting

process. The output includes an extensive nonlinear statistical analysis of the differences between measurement and theory in addition to the parameter results $p \pm \delta p^k$. The statistical analysis decisions are based on a nonlinear generalization of the Student and Fisher theory. Assessments of the quality of the measurements and the relevance of the theory are made in addition to the adequacy of agreement between measurement and theory. Graphical representation of enormous output detail is also available in a magnification chosen by the user.

The general statistical analysis capability of Program LIKELY has been adapted⁽³⁴⁾ to the solution of specific analysis problems. One of these DBUFIT-I, a double precision burnup fitting code,⁽³⁵⁾ has been specifically developed for obtaining the integral cross section information from isotopic transmutation data. The DBUFIT-I code is an improvement over the DUBLIK code⁽³⁴⁾ which had previously been used for the least squares analysis of burnup data.

APPLICATION USING Al-Pu ELEMENTS

Samples of Al-Pu from fuel elements which have been irradiated in the PRTR⁽³⁶⁾ have been destructively analyzed to determine plutonium depletions and isotopic compositions. The elements were 19-rod clusters which initially contained two plutonium compositions. One type of element contained 88 in. long rods of an alloy Al-2 wt% Ni - 1.82 wt% Pu.⁽⁶⁾ Each element contained 268 g plutonium and the isotopic composition before irradiation was 93.28/6.25/0.457/0.0178 at% $^{239}\text{Pu}/^{240}\text{Pu}/^{241}\text{Pu}/^{242}\text{Pu}$,

respectively. The second type contained an alloy Al-2.1 wt% Ni - 2.60 wt% Pu which had an isotopic composition before irradiation of 81.06/16.45/2.29/0.20 and 376 g plutonium per cluster.⁽⁷⁾ The rods of both elements were clad with 0.035 in. Zircaloy-2.

The PRTR (a vertical pressure tube, heavy water moderated and cooled reactor) was operated at a thermal power of 70 MW. The inlet and outlet temperatures were 470 and 530°F or 125 and 150°F for the coolant or moderator, respectively. The fuel elements were located on an 8 in. pitch with fuel elements different from the experimental element. However, the experimental elements were surrounded by other elements of the same composition and exposure history.

The fuel elements were removed from the reactor at approximately equal increments of exposure up to a maximum depletion of about 50% of the initial plutonium. Detailed analyses to obtain burnup data were carried out for one rod from each of two rings and the center rod of the clusters. The data describes the variation of the concentration of plutonium isotopes as the plutonium is being depleted.^(6,7) The total irradiation exposure is computed from the depletion data and effective cross section ratios are derived from the atom ratios and fuel depletion.

Data from samples having the same initial fuel composition and irradiated in approximately the same nuclear surroundings have been analyzed together. This results in four groups of data:

- 1.82 wt% Pu in Al(6.25% ^{240}Pu) from rods of the outer ring of the cluster (Lx0)

- 1.82 wt% Pu in Al (6.25% ^{240}Pu) from rods of the inner ring and center rod of the cluster (LxMC)
- 2.6 wt% Pu in Al (16.45% ^{240}Pu) from rods of the outer ring of the cluster (Hx0)
- 2.6 wt% Pu in Al (16.45% ^{240}Pu) from rods of the inner ring and center rod of cluster (HxMC).

If it is assumed that the samples analyzed in each group have been irradiated in neutron spectra common to that group, then it can be concluded that the fuel composition depends only upon initial fuel composition and the exposure it received.

In many reactors, the items which perturb the neutron spectrum in the course of an extended irradiation are control rods, burnable poisons, isotopic fuel transformations, and leakage. Other factors, such as temperature, type and density of the moderator, and structural components which affect the spectrum remain relatively constant during the irradiation. In the absence of burnable poisons and in fuel locations sufficiently removed from the control rods, only the fuel composition alters the local spectrum and hence the flux averaged cross sections of the fuel. These in turn govern the incremental fuel changes and suggest that the fuel composition evolves in a manner amenable to the differential or integrated equation method of analysis.

The unique relationship between fuel composition and exposure combined with the fact that the exposure varies along a rod has the advantage of allowing many data points to be gathered from the same rod. In this

way the number of rods and reactor shutdowns required to obtain a given number of samples is reduced. Samples cut from the ends of the rod have been omitted from analysis because the perturbation of the flux spectrum at these positions by flux leakage effects is suspected to cause significant effects.

Samples from rods in similar positions of different elements have been analyzed in one group since they also have been irradiated in common spectra. Even though some adjacent PRTR positions were loaded with different fuel elements they do not greatly influence the spectrum of the experimental elements since neutrons originating in one element are thermalized by the time they reach another. Again, fuel composition changes of each element are solely dependent upon initial fuel composition and exposure of that element.

Differential Method

Expansion of Equations (5) for the plutonium system results in three equations containing five unknown ratios of effective cross sections. Solving for the cross section ratios allow three of them to be determined from the experimental data and equations (6) and (8). The effective cross section ratios

$$\hat{\sigma}_c^{49}/\hat{\sigma}_a^{49} = \epsilon; \hat{\sigma}_a^{40}/\hat{\sigma}_a^{49} = E; \hat{\sigma}_a^{42}/\hat{\sigma}_a^{49} = D \quad (19)$$

$$(\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49})(1 + \lambda^{41}/\hat{\sigma}_a^{41}\phi) = C; \text{ and} \quad (20)$$

$$(\hat{\sigma}_c^{41}/\hat{\sigma}_a^{41})(1 + \lambda^{41}/\hat{\sigma}_a^{41}\phi)^{-1} = \gamma \quad (21)$$

All radioactive decay of the isotopes are considered negligible except for ^{241}Pu (i.e., $\lambda^{41} \neq 0$) and $\hat{\sigma}_c^{40} = \hat{\sigma}_a^{40}$. An average value of $\lambda^{41}/\hat{\sigma}_a^{41}\phi = 0.050 \pm 0.025$ is assigned to each group of data on the basis of experimental and calculated information. A large uncertainty is assigned to the group average value since it varies widely from one data point to another.

Since there are more unknowns than equations two other equations must be established before numerical values can be obtained for the ratios. The ratio D is relatively small and could be set equal to zero; however, a calculated value of $D = 0.07$ is a better approximation. Another calculated value $C = 1.20$ is used because of the greater confidence placed in its calculation because of the nature of the basic cross sections and the determination that an incorrect value would have a smaller effect on the remaining cross section ratios.

The relations between the ratios and the data are

$$\epsilon = - \left[\Delta N^{40} + \Delta N^{41} - CI_1 \right] / \Delta N^{49} \quad (22)$$

$$C = - \left[\Delta N^{41} - CI_1 \right] / \int_a^b (N^{40}/N^{49}) dN^{49}; \quad (23)$$

$$\gamma = - \left[\Delta N^{42} - DI_2 \right] / I_1 C \quad (24)$$

where

$$I_1 = \int_a^b (N^{41}/N^{49}) dN^{49}$$

and

$$I_2 = \int_a^b (N^{42}/N^{49}) dN^{49}.$$

The ΔN^i are changes in plutonium concentrations in at/b-cm and ϵ , E, C, D, and γ are assumed constant for a particular burnup interval (b-a). The ratios N^i/N^{49} are represented by functions of the form given by equation (6). The parameters A_k of these equations are determined from the experimental data by the method of least squares.^b This in turn allows values of the ΔN^i to be obtained from equation (8). Finally, the ratios ϵ , E and γ are determined as a function of plutonium depletion. In the process the value of $\hat{\alpha}^{49} = \hat{\sigma}_C^{49}/\hat{\sigma}_F^{49}$ and $\hat{\alpha}^{41} = \hat{\sigma}_C^{41}/\hat{\sigma}_F^{41}$ is determined. The

$$\hat{\alpha}^{49} = \epsilon/(1-\epsilon) \text{ and } \hat{\alpha}^{41} = \gamma/(1-\gamma) \quad (25)$$

where

$$\gamma = \gamma(1 + \lambda^{41}/\hat{\sigma}^{41}\phi).$$

The values obtained for the ratios of effective cross sections are summarized in Table II as a function of depletion for rods of the outer ring of the cluster (Lx0) and for rods of the inner ring and center rod of the cluster (LxMC). Values of $\hat{\alpha}^{49}$, $\hat{\alpha}^{41}$, and $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ were obtained for five equal subintervals between 1.5 and 48% depletion. Values of $\hat{\alpha}^{49}$ and $\hat{\alpha}^{41}$ were constant within the experimental uncertainty and the averages for the total interval confirm the values obtained by the integrated equation method (Table V). The values of $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ were not constant and agree with the values obtained in the integrated equation analysis and corroborated the shape used for the ratio^c (Figure 4).

TABLE II

CROSS SECTION RATIO VALUES AND THEIR ONE STANDARD DEVIATIONS
FOR Al-Pu CONTAINING INITIALLY 6% ^{240}Pu

Cross Section Ratio	Percent Depletion Interval					
	1.5 to 10.2	10.2 to 19.5	19.5 to 28.8	28.8 to 38.2	38.2 to 48.2	
$\hat{\sigma}_c^{41}/\hat{\sigma}_a^{49}$	Lx0	0.289 ± 0.011	0.296 ± 0.008	0.305 ± 0.007	0.307 ± 0.011	0.312 ± 0.019
	LxMC	0.296 ± 0.010	0.295 ± 0.008	0.302 ± 0.008	0.314 ± 0.007	0.311 ± 0.012
$\hat{\alpha}^{41}$	Lx0	0.336 ± 0.022	0.347 ± 0.016	0.360 ± 0.016	0.365 ± 0.023	0.373 ± 0.040
	LxMC	0.346 ± 0.021	0.345 ± 0.016	0.356 ± 0.017	0.376 ± 0.016	0.370 ± 0.026
$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$	Lx0	0.507 ± 0.014	0.451 ± 0.010	0.437 ± 0.009	0.425 ± 0.014	0.391 ± 0.024
	LxMC	0.456 ± 0.012	0.404 ± 0.009	0.390 ± 0.008	0.382 ± 0.010	0.350 ± 0.013
$\hat{\sigma}_c^{49}/\hat{\sigma}_a^{49}$	Lx0	0.300 ± 0.009	0.305 ± 0.008	0.310 ± 0.009	0.313 ± 0.010	0.308 ± 0.020
	LxMC	0.299 ± 0.009	0.301 ± 0.009	0.305 ± 0.009	0.310 ± 0.009	0.319 ± 0.015
$\hat{\alpha}^{49}$	Lx0	0.429 ± 0.018	0.438 ± 0.017	0.450 ± 0.018	0.455 ± 0.022	0.446 ± 0.042
	LxMC	0.426 ± 0.018	0.430 ± 0.017	0.438 ± 0.018	0.449 ± 0.020	0.468 ± 0.032

Integrated Equations Method

The same definitions and assumptions of the differential method are used in the expansion of equation (4) and the integrated equation method. The approximation that $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ equals a constant E is no longer valid since the burnup increment is not small. Therefore another relationship is sought which will still allow analytical solutions to be obtained for the differential equations. The choice of

$$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49} = A + B/N^{40} \quad (26)$$

where A and B are constants adequately meets the needs of the Al-Pu analysis. The other ratios are assumed constant as in equations (19-21).

Under these conditions, equations (4) are integrated by the standard technique of transforming the equations to exact differentials with appropriate integrating factors. The solutions are

$$N^{40} + B/A + N^{49} \epsilon(1-A)^{-1} - K_0(N^{49})^A = 0 \quad (27)$$

$$N^{41} - \epsilon A(1-A)^{-1}(1-C)^{-1} N^{49} + AK_0(A-C)^{-1}(N^{49})^A - K_1(N^{49})^C = 0 \quad (28)$$

$$\gamma^{-1}N^{42} + \epsilon AC(1-A)^{-1}(1-C)^{-1}(1-D)^{-1} N^{49} - ACK_0(A-C)^{-1}(A-D)^{-1}(N^{49})^A + CK_1(C-D)^{-1}(N^{49})^C - K_2(N^{49})^D = 0 \quad (29)$$

The K_i are determined from equations (27), (28), and (29) when the initial concentrations N_0^i are substituted for the N^i . The other parameters are defined by equations (19), (20), and (21). Equations (27), (28), and (29) are precisely the $q(y,p)$ equations required by the least squares fitting analysis. The fitted parameters p are the K_i , ϵ , A, C, γ , and D while the variables y are the isotopic concentrations N^i . The variance-covariance matrix (equation 12) for each data point is calculated from random measurement errors using standard techniques of propagation of errors. The cross section ratio D is too small to be

determined from the experimental data. Therefore, the calculated value of 0.07 is used as before. After a satisfactory fit has been obtained with the equations, the fraction β of plutonium atoms destroyed can be calculated. Neglecting the effects of ^{242}Pu neutron capture, β is the sum of ^{239}Pu and ^{241}Pu atoms fissioned and ^{241}Pu atoms decayed divided by the initial plutonium content or

$$\beta = \frac{(1-\epsilon)(N_0^{49}-N^{49}) + (1-\gamma)\gamma^{-1}(N^{42}-N_0^{42})}{N_0^{49}+N_0^{40}+N_0^{41}+N_0^{42}} \quad (30)$$

where the zero subscripts denote initial values. In the actual analysis the more complicated expression is used which accounts for ^{242}Pu neutron capture.

In principle, a generalized least squares fit to the experimental data using equations (27), (28), and (29) will determine all of the unknown parameters included in the equations. In practice, the various sets of data for the Al-Pu may yield more than one solution unless one of the cross section ratios is preset and held fixed during the iterative fitting process. For this purpose $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$ is chosen as in the differential method.

Graphical results of generalized least squares fits are shown in Fig. 2 for the 1.8 wt% Pu, outer ring case (Lx0). The three isotopic curves are the specific projections of the generalized curve of Fig. 1 onto the appropriate planes. Because of the small distance between one standard deviation limits on each curve, the area between them has been shaded in for purposes of clarity. The experimental data have been

Figure 2. Graphical Results of Generalized Least Squares Fits for 1.8 wt% Pu.

plotted showing the one standard deviation measurement error flags. Error flags for most points fail to show up because they are smaller than the mark indicating the data point. The plutonium depletion β predicted from the best fitting parameters and equation (3) is shown in Fig. 3. The β values determined experimentally from ^{137}Cs data are also plotted for comparison. The curves for the other three cases (LxMC, HxO, and HxMC) are quite similar to the ones shown and the goodness of the fits are about the same.

The cross section ratios obtained from the least squares fits are presented in Table III for all the Al-Pu data. A uniformity of the ratios for the four cases is expected for $\hat{\sigma}_a^{41}$ and $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$. In the thermal energy region $\alpha^{41}(E)$ changes little with neutron energy and spectrum changes should not change its effective value. The $\sigma_a^{49}(E)$ and $\sigma_a^{41}(E)$ have similar shapes and as a result $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$ is relatively unaffected by neutron spectrum changes. On the other hand, $\alpha^{49}(E)$ does vary appreciably with neutron energy in the thermal region and since the experimental values are approximately the same it is concluded that the thermal neutron spectra of the four cases are not appreciably different.

The ^{241}Pu decay correction term $\lambda^{41}/\hat{\sigma}_a^{41}\bar{\phi}$ required to determine $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$ from the least squares value C has been calculated from the heat released by the element. Heat flow is monitored for each PRTR element and $\bar{\phi}$ is calculated using the formula $H(\text{MWd}) = kV\bar{\Sigma}_{\text{fis}}\bar{\phi}\Delta t$. The $k = 3.91 \times 10^{-22}$ MWd/fis and Δt is the time in seconds during which the element was irradiated. The $\bar{\Sigma}_{\text{fis}}$ and $\hat{\sigma}_a^{41}$ were computed values.

PLUTONIUM DEPLETION FOR LXO FUEL

Figure 3. The Plutonium Depletion for 1.8 wt% Pu as a Function of ^{239}Pu Concentration.

TABLE III

CROSS SECTION RATIOS FROM INTEGRATED EQUATION METHOD

The values are obtained by keeping $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49} = 1.1429$, $\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49} = 0.07$ and $\lambda^{41}/\hat{\sigma}_a^{41}\bar{\phi} = 0.05 \pm 0.025$ during the fitting process. Numbers in parentheses are one standard deviation uncertainties obtained from the fitting process.

Fuel	Highest Fuel Depletion (%)	$\hat{\alpha}^{49}$	$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$		$\hat{\alpha}^{41}$
			A	B x 10 ⁵	
LxO	50.4	0.4391 (0.0032)	0.3097	0.2153	0.351 (0.013)
LxMC	48.0	0.4361 (0.0032)	0.2673	0.2102	0.355 (0.013)
HxO	51.2	0.4421 (0.0026)	0.2043	0.7305	0.342 (0.012)
HxMC	48.1	0.4465 (0.0025)	0.1751	0.7133	0.339 (0.011)

Finally, $\lambda^{41} = 1.655 \times 10^{-9} \text{ sec}^{-1}$ ($T_{1/2} = 13.3 \text{ yrs}$ for ^{241}Pu) completes the information necessary for the correction.^(c) Values of $\bar{\phi}$ vary widely for individual elements as well as for individual samples taken from within these elements. Also, $\bar{\Sigma}_{\text{fis}}$ and $\hat{\sigma}^{41}$ change appreciably during the irradiation of the fuel. Consequently, the choice of a single value for $\lambda^{41}/\hat{\sigma}_a^{41}\bar{\phi}$ for all data points of a given case is quite uncertain and has been assigned an error of 50%. Once $\lambda^{41}/\hat{\sigma}_a^{41}\bar{\phi}$ has been estimated, its effect on the values of $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$ and $\hat{\alpha}^{41}$ can be computed.

The values of $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ are plotted in Fig. 4 as a function of the ^{240}Pu atom concentration while the ^{239}Pu was depleted to ~50% of its original value. Also plotted in the figure are values obtained from the differential methods of analysis. The solid curves are defined by the A and B values obtained in the simultaneous least squares fit and the bounds of the curves are the standard deviations determined from the fits. The crosses plotted in the figure were obtained by the differential analysis method which allows the variation of all the effective cross section ratios from the data. The agreement between the crosses and the curves lend credence to the shape assumed for the simultaneous fit.

Figure 4. Values of $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ as a Function of ^{240}Pu Atom Concentration

USE OF THE DATA FOR TESTING CALCULATIONS

In reactor neutronics calculations, the initial step is to calculate the multigroup neutron spectrum for the basic representative sub-region or unit cell of the reactor. The neutron spectrum within this unit cell is usually computed using digital computer programs which solve the Boltzmann equation for neutron transport. Multigroup (the order of 100 groups or more) values of the neutron cross sections are averaged over this neutron spectrum to reduce the multigroup values to few group values. Traditionally, the calculation of the neutron spectrum is divided into non-thermal groups (10^7 eV to thermal cutoff) and a thermal group (thermal cutoff to 0.0 eV).

In this section we show how the cross section values obtained with the unit cell codes are used to obtain the one group values needed for comparison to the ratios deduced from the fit to the isotopic data. We then illustrate the use of the deduced cross section ratios and the calculated values in verification of burnup calculations. In the process the theoretical model employed and the results obtained are described.

Calculational Approach

An approach which is simple in practice and expected to be accurate in principle is used to calculate the cross section ratios for comparison with the values deduced from the experimental data. The approach is based upon using the measured isotopic concentrations for the fissile and fertile nuclides in the fuel in calculations of the neutron spectrum in a unit cell of the reactor. Thus, it is assumed that the measured fertile and fissile nuclides characterize the neutron spectrum within a unit cell and that fission products do not perturb the spectrum significantly.

The spectrum averaged values, called here effective values, are defined according to equation (2)

$$\bar{\sigma}^{i,j} = \frac{\int_{V_j} \int_0^{10^7 \text{ eV}} \sigma^i(\vec{r}, E) \phi(\vec{r}, E) dE d\vec{r}}{\int_{V_j} \int_0^{10^7 \text{ eV}} \phi(\vec{r}, E) dE d\vec{r}} \quad (31)$$

where the spatial integration is over the fuel volume V_j of interest.

The neutron spectrum within a unit cell is usually characterized into thermal and non-thermal neutron energy groups. Thus, the multigroup values for each isotope must be reduced to spectral average values for the thermal and non-thermal energy groups. Separating the energy integrals in numerator and denominator into non-thermal (E_c to 10^7 eV) and thermal (0.0 to E_c) terms and assuming the spatial integral of the nonthermal flux above E_c is constant allows equation (31) to be written as

$$\bar{\sigma}^{i,j} = \frac{\bar{\sigma}_1^i \phi_1 + \int_{V_j} \int_{0.0}^{E_c} \sigma^i(\vec{r}, E) \phi(\vec{r}, E) dE d\vec{r}}{\phi_1 + \int_{V_j} \int_{0.0}^{E_c} \phi(\vec{r}, E) dE d\vec{r}} \quad (32)$$

where subscript 1 refers to the nonthermal neutron energy group and E_c is the energy boundary between the thermal and nonthermal groups. The second term of the numerator is spatially constant and can be written as the product of the average thermal cross section, $\bar{\sigma}_2^{i,j}$ and the total thermal neutron flux ϕ_2^j in the fuel region of interest. Subscript 2 refers to the thermal neutron energy group. The second term in the denominator is just the total thermal neutron flux ϕ_2^j in fuel region j. Thus, equation (32) becomes

$$\hat{\sigma}^{i,j} = (\bar{\sigma}_1^{i,j} \phi_1^j + \bar{\sigma}_2^{i,j} \phi_2^j) / (\phi_1^j + \phi_2^j) . \quad (33)$$

The fluxes ϕ_1^j and ϕ_2^j are normalized assuming that all neutrons scattered from group 1 to group 2 are absorbed in group 2. Thus

$$\phi_1^c \bar{\Sigma}_{1 \rightarrow 2}^c = \phi_2^c \bar{\Sigma}_{a2}^c , \quad (34)$$

where ϕ_1^c and ϕ_2^c are the total flux in the cell for group 1 and 2. Now ϕ^c is related to the total flux in the fuel by

$$\phi_1^j = \phi_1^c (V^j \bar{\phi}_1^j) / (V^c \bar{\phi}_1^c) \text{ and} \quad (35)$$

$$\phi_2^j = \phi_2^c (V^j \bar{\phi}_2^j) / (V^c \bar{\phi}_2^c) \quad (36)$$

where V refers again to volume and $\bar{\phi}$ is the average flux. Assuming $\phi_1^j / \bar{\phi}_1^j = 1$, substituting equations (35) and (36) in equation (34) and solving for the ratio ϕ_1^j / ϕ_2^j and then using this normalizing condition in equation (33), the expression for the effective cross section becomes

$$\hat{\sigma}^{i,j} = \frac{(\bar{\sigma}_1^{i,j} \bar{\Sigma}_{a2}^C / \bar{\Sigma}_{1 \rightarrow 2}^C) + (\bar{\sigma}_2^{i,j} \bar{\phi}_2^j / \bar{\phi}_2^C)}{(\bar{\Sigma}_{2a}^C / \bar{\Sigma}_{1 \rightarrow 2}^C) + (\bar{\phi}_2^j / \bar{\phi}_2^C)} \quad (37)$$

The quantities $\bar{\sigma}_1^j$ and $\bar{\Sigma}_{1 \rightarrow 2}^C$ are obtained from cell calculations using a code which calculates the slowing down of neutrons and all other quantities are obtained from thermalization calculations. The effective cross sections as calculated with equation (37) for each isotope are used to obtain the ratios needed for comparison to the values deduced from the fit to the isotopic data.

Theory-Experiment Comparison

The cross section ratios which were deduced from the Al-Pu fuel irradiations in the PRTR are used to illustrate how these data can help verify the accuracy of burnup calculations. The measured isotopic concentrations at pre-selected exposures are used as input to cell codes for unit cell calculations to obtain ratios of effective cross sections. These ratios are compared to the values deduced from the experimental data to provide a basis for verifying the accuracy of the calculated results. Similar comparisons using results from calculations which use calculated isotopic concentrations is an alternate approach which can provide additional information.

Theoretical Methods

The theoretical methods used at BNW for this study are based on approximate solutions of the neutron transport equation. The neutron distributions in space and energy within a single cell of the reactor were computed using the multigroup transport theory codes, HRG⁽³⁷⁾ and

THERMOS.⁽³⁸⁾ All basic neutron cross sections for these cell codes were obtained from the Battelle-Northwest Master Library (BNWML).⁽³⁹⁾ The thermal neutron cross sections for the fissile nuclides were normalized to the 2200 m/sec values given in the 1965 International Atomic Energy (IAEA) Evaluation conducted by Westcott, et. al.⁽⁴⁰⁾

The behavior of neutrons slowing down to thermal energies is computed using the HRG code. The time independent Boltzmann equation is solved with HRG for isotropic sources of neutrons in the B-1 or P-1 approximations. The neutron flux and current spectra are computed assuming 68 groups of neutrons with a constant group width of lethargy = 0.25. Corrections for heterogeneity, Doppler broadening, and leakage are included. The boundary dividing the thermal and non-thermal energy groups (E_c of equation 32) was chosen as 0.683 eV. The multigroup cross sections were reduced to a one group value for the energy region from 0.683 to 10^7 eV.

The thermal neutron spectrum is computed using the THERMOS code. The version of this code in use at PNL for this study was flexible as to the number of energy groups (up to 30), space points (up to 30) and mixtures (up to 8) such that the code can be tailored to the specific problem to be solved. The thirty group cross sections for each isotope were spectral averaged to obtain one group values for the energy range from 0.0 to 0.683 eV.

Neutron Spectrum Computations

The neutron spectrum is computed at various exposures utilizing the experimental atom concentration of the plutonium isotopes. The

average physical temperatures of the fuel, coolant, and moderator were assumed to be 343, 260, and 59°C, respectively. These are the average inferred fuel temperature during irradiation and the average of the observed inlet and outlet temperatures for the moderator and coolant for the positions in which these Al-Pu fuels were irradiated in the PRTR.

The slowing down of neutrons is computed using the HRG code. The options utilized in the current study are the P-1 approximation for zero leakage, the ^{239}Pu fission spectra, and Doppler broadening and spatial self shielding corrections for ^{239}Pu , ^{240}Pu , and ^{241}Pu .

Spatial corrections are based upon the methods developed by Nordheim.⁽⁴¹⁾ The mean chord length ($\bar{l}$) for the cluster was obtained from the relationship $\bar{l} = 4V/S_{\text{eff}}$ where V is the volume enclosed by a rubber band stretched around the cluster and $S_{\text{eff}} = 1.23 \times \text{rubber band surface area}$. The admixed moderator scattering cross section per absorber atom was computed for a single rod of the 19-rod cluster, (i.e., Al as the admixed moderator in the fuel). This procedure for obtaining the spatial correction parameters evolved empirically from analytical correlations with Hellstrand's experiments for 19-rod clusters.⁽⁴²⁾

The thermal neutron spectrum is computed using the THERMOS code. Thirty neutron energy groups were used to describe events occurring below 0.683 eV. A concentric cylinder model of the cluster obtained by preserving atom concentrations and volumes was used. The cell is described in seven regions with 30 space points. The Honeck-Nelkin kernel was used for D_2O ⁽⁴³⁾ with an approximate correction to the kernel to account

for anisotropy. ⁽⁴⁴⁾ The scatter-transfer from all other cell materials were computed with the Brown-St. John Model. ⁽⁴⁵⁾ Reflecting cell boundary conditions were also utilized.

To simplify the calculation, it was assumed that the neutron spectrum in the fuel was, to the first order, sensitive to only the concentration of fissile and fertile nuclides. That is, the fission product inventory and the presence of neutron leakage affect mainly the neutron population and their affect on the neutron spectrum is negligible.

Comparison of Results

A set of ratios obtained from results of cell calculations and equation (15) are compared in Table IV to values obtained from the least squares analysis of experimental data using Program DUBLIK in Table IV. The calculated values of $\hat{\alpha}^{49}$ and $\hat{\alpha}^{41}$ are within one standard deviation of the experimental values and lower by approximately 4-1/2 and 9%, respectively. The values deduced in the least squares analysis show slight variations between the outer 12 rods and the inner six rods whereas the calculated values do not. Because of the disagreement in this variation, it appears that both the basic energy self shielding and the spatial self shielding is being calculated incorrectly.

The calculated values of $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ are outside the standard deviations of the experimental values. However, a discrepancy was expected in these ratios since the calculation of resonance absorption in ^{240}Pu was made assuming a homogeneous cell. The experimental data itself suggest that spatial effects are important in calculating resonance absorption in ^{240}Pu as shown by the

TABLE IV

COMPARISON OF CALCULATED RATIOS OF EFFECTIVE CROSS SECTIONS
TO VALUES OBTAINED FROM LEAST SQUARES ANALYSIS
OF EXPERIMENTAL DATA FOR Lx Pu-A1 CLUSTERS

Fractional Burnup	Calculated			Least Squares Analysis	
	Ratio	Outer 12 Rods of Cluster	Inner 6 Rods of Cluster	Outer 12 Rods	Inner 7 Rods
0.003	$\hat{\alpha}^{49}$	0.458	0.458	0.439 ± 0.019	0.436 ± 0.019
	$\hat{\alpha}^{41}$	0.384	0.384	0.357 ± 0.030	0.355 ± 0.030
	$\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$	1.189	1.199	1.143 (Fixed)	1.143 (Fixed)
	$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$	0.468	0.503	0.581 ± 0.016	0.531 ± 0.016
	$\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49}$	0.094	0.104	0.07 (Fixed)	0.07 (Fixed)
0.1238	$\hat{\alpha}^{49}$	0.456	0.457		
	$\hat{\alpha}^{41}$	0.334	0.384		
	$\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$	1.192	1.199		
	$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$	0.339	0.409	0.466 ± 0.015	0.420 ± 0.015
	$\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49}$	0.085	0.092		
0.2351	$\hat{\alpha}^{49}$	0.454	0.456		
	$\hat{\alpha}^{41}$	0.384	0.384		
	$\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$	1.196	1.199		
	$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$	0.350	0.362	0.427 ± 0.015	0.382 ± 0.015
	$\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49}$	0.077	0.084		
0.3639	$\hat{\alpha}^{49}$	0.452	0.454		
	$\hat{\alpha}^{41}$	0.384	0.384		
	$\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$	1.201	1.201		
	$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$	0.320	0.326	0.405 ± 0.014	0.360 ± 0.014
	$\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49}$	0.067	0.070		
0.4852	$\hat{\alpha}^{49}$	0.449	0.452		
	$\hat{\alpha}^{41}$	0.384	0.384		
	$\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$	1.206	1.204		
	$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$	0.300	0.302	0.398 ± 0.014	0.352 ± 0.014
	$\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49}$	0.059	0.061		

variation of the ratio $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ between the outer 12 rods and the inner six rods of the cluster. Since the other three ratios are fixed in the least squares analysis no information could be obtained from their comparison.

A simple way to resolve the $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ discrepancies would be to force the data to agree via the spatial correction (mean chord length, $\bar{\ell}$) term in the slowing down calculation of the HRG code. For example, calculations of the ratio $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ for different mean chord lengths $\bar{\ell}$ resulted in the conclusion that a value of $\bar{\ell}$ between 2 and 3 instead of the value 6.95 which was used would provide a cross section ratio in reasonable agreement with experiment. The Bell⁽⁴⁶⁾ prescription is found to give $\bar{\ell} = 2.9$ when applied as follows:

$$\bar{\ell} = \frac{\ell_0 + V_0/V_1}{\hat{\Sigma}_1}$$

where ℓ_0 = diameter of fuel in a rod of the cluster,

V_0 = volume of fuel in the cluster,

V_1 = volume enclosed by a rubber band enriching the cluster, and

$\hat{\Sigma}_1$ = effective epithermal neutron cross section when the cladding and D₂O coolant are homogenized.

Hence, if the criterion were to match calculation and experiment for the $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ the Bell recipe would provide a reasonable match for this set of experiments.

As mentioned above, the effects of neutron leakage and the presence of fission products (gaseous and stable nuclides) have been neglected in the calculations. The impact of neglecting the leakage effect was evaluated by performing a slowing down calculation (P-1 approximation in the HRG code) using the geometrical buckling, $B_g^2 \approx 5 \text{ m}^{-2}$, for the PRTR loading as the leakage factor in the calculation (assuming leakage does not affect the thermal cross section averages). The cross sections from this calculation are compared in Table V to those obtained where leakage was neglected completely. The effective absorption cross sections for ^{239}Pu and ^{240}Pu change by slightly less than 2% but the ratios are unaffected when leakage is included in the calculation. In order to observe changes in the ratios, buckling values larger than 50 m^{-2} are required. Similar calculations of the effect of including thermal neutron leakage show that the local buckling of the fuel zone where the samples were irradiated must be larger than the geometrical buckling of the reactor to introduce errors in the analysis sufficient to account for the observed discrepancy. A result of a two dimensional diffusion theory reactor calculation for the loading in which the data were collected shows that the gradient across the sampling zone is negligible. Thus, the assumption that leakage does not affect the thermal cross section averages appears valid.

TABLE V

THE EFFECT OF LEAKAGE ON EFFECTIVE CROSS SECTIONS AND RATIOS

Fractional Burnup	Effective Cross Sections, $\hat{\sigma}$ (barns)				Ratios			
	$\hat{\sigma}_a^{49}$		$\hat{\sigma}_a^{40}$		$\hat{\alpha}_a^{239}$		$\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$	
	No Leakage	Leakage	No Leakage	Leakage	No Leakage	Leakage	No Leakage	Leakage
0.003	497.9	489.7	233.0	229.0	0.458	0.457	0.468	0.468
0.2351	567.4	559.4	198.3	195.5	0.454	0.454	0.350	0.349

A calculation of the neutron spectrum was made with fission products included in the fuel of the cell. The concentrations of fission products were taken from results of an earlier burnup calculation⁽⁴⁷⁾ performed for this same cell. The fission products ^{135}Xe , ^{149}Sm , ^{151}Sm , ^{155}Eu , ^{155}Gd , and four pseudo elements⁽⁴⁸⁾ were assumed to be present. The cross sections calculated using the spectrum which include fission products are compared in Table VI to those obtained when fission products were neglected. The effective cross section for ^{239}Pu changes by about 2-3% whereas the value for ^{240}Pu is relatively unaffected indicating a change mainly in the thermal neutron energy component of $\hat{\sigma}$. The ratio $\hat{\alpha}^{49}$ is insensitive to the effect of fission products. The ratio $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ increases by about 1 to 4% between 25 and 0.3% depletion, respectively, when the fission products are considered.

In summary, the ratios $\hat{\alpha}^{49}$, $\hat{\alpha}^{41}$, and $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ are all different from the values deduced in the fit. However, $\hat{\alpha}^{49}$ and $\hat{\alpha}^{41}$ are within one standard deviation. The neglect of the fission products and neutron leakage in the spectrum calculation does not introduce significant error in the calculated results. Therefore, assuming that the method of deducing the values from the isotopic data is correct, the deviation between the calculated and deduced values must be caused by the calculated results and occurs because of inaccurate theoretical models and basic neutron cross section data.

Examination of Potential Causes of the Discrepancy

The accuracy of calculating fuel burnup behavior is usually evaluated by using the initial fuel concentration in a time dependent calculation of the flux and spectrum average cross sections. In the evaluation a

TABLE VI

THE EFFECT OF FISSION PRODUCTS ON EFFECTIVE CROSS SECTIONS AND RATIOS

Fractional Burnup	Effective Cross Sections, σ (b.)				Ratios			
	σ_a^{49}		σ_a^{40}		σ_a^{49}		$\sigma_a^{40}/\sigma_a^{49}$	
	No F.P.*	F.P.	No F.P.	F.P.	No F.P.	F.P.	No F.P.	F.P.
0.003	497.9	481.5	233.0	233.9	0.458	0.459	0.468	0.486
0.2351	567.4	557.1	198.3	197.3	0.454	0.456	0.350	0.354

*F.P. meaning fission products.

determination is made concerning how well the isotopic variations with exposure are reproduced. Each isotopic concentration is compared separately and usually various degrees of accuracy are obtained. Correlations of the effective cross section ratios are believed to be a more stringent test than the isotopic concentration. A set of ratios are obtained which result in an equally good fit to all of the measured isotopic data. In addition, they allow a separate evaluation of cross sections and theoretical models since the uncertainty of calculating the isotopic concentrations can be removed by making a comparison independent of the transmutation calculation. This is accomplished by including the measured concentrations in the calculation. However, the method of deducing the cross section ratios must be correct in order to obtain valid conclusions about the calculations.

The results of a study by Page ⁽⁴⁷⁾ using the same Al-Pu data have shown that when agreement is obtained between measured and calculated isotopic concentrations the calculated and deduced cross section ratios were also in agreement. Based upon the results of that study it is expected that the values of deduced cross section ratios in Table IV are valid.

The results from Reference 47 also validate the calculational approach presented in this paper. In addition, the results of a previous study based upon our calculational approach ⁽⁴⁹⁾ agree with those obtained by Page when the basic neutron cross sections ^(50, 51) are identical in both calculations.

The calculated values of $\hat{\alpha}^{49}$ and $\hat{\alpha}^{41}$ obtained from each study were in good agreement ($\pm 1\%$) whereas the values of $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ obtained by Page were different from other values obtained in our study. The principal reason for the disagreement in this ratio is that Page used a one-dimensional model to calculate the neutron spectra in the vicinity of the large ^{240}Pu resonance at 1.056 eV. This one-dimensional representation provides a better calculation of the neutron spectra in this energy region and therefore, should give better values for the spectrum averaged cross section for ^{240}Pu .

Improvements have been made to the HRG and THERMOS cell codes and are reported as HRG-3 and BRT-I, respectively. ^(52,53) In addition, new evaluations of cross sections were made available after these studies were completed. The need to modify the cell codes grew out of these results and the results of correlations of other calculated and measured reactor parameters. ⁽⁵⁴⁾ The advent of the Evaluated Nuclear Data File (ENDF/B) has provided improved estimates of the neutron cross sections. The 1965 IAEA evaluation ⁽⁴⁰⁾ has been updated ⁽⁵⁵⁾ and included in Version II of the ENDF/B file as the basis of normalization of the fissile nuclides. ⁽⁵⁶⁾

The modifications incorporated in HRG-3 affect the calculations of the resonance integral by adapting the method of Adler, Hinman, and Nordheim to an intermediate resonance formulation, by distributing the contribution of broad resonances over several fine groups, and by correcting the epithermal transfer cross sections in water to allow for upscattering by hydrogen. These modifications result in appreciable increases in the calculated nonthermal cross sections of ^{240}Pu and ^{242}Pu but give only

small changes in the nonthermal cross sections of ^{239}Pu and ^{241}Pu .

However, the resonance allocation change increases the absorption whereas the intermediate resonance formulation decreases the absorption rate with the net result that these changes tend to cancel. The additional upscattering by hydrogen is negligible for the D_2O moderated and cooled PRTR fuel. The calculated ratio $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ and $\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49}$ therefore is expected to increase by a few percent with these modifications, while the ratios $\hat{\alpha}^{49}$, $\hat{\alpha}^{41}$, and $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$ would remain essentially unchanged. The modifications incorporated in the BRT-I code include generalized boundary conditions, an improved current approximation, and more practical edits. None of these changes are expected to significantly modify the results of this study.

The resonance integrals for infinite dilution and the 2200 m/sec values for the cross sections used in this study for the plutonium isotopes are summarized in Table VIII. Also shown are the values contained in Version II of the ENDF/B file. ⁽⁵⁷⁾

The difference in the absorption cross sections at 2200 m/sec is not significant enough to cause a spectrum change. Therefore, the difference in the effective values would be proportional ⁽⁵⁸⁾ to those of the 2200 m/sec values for ^{239}Pu and ^{241}Pu . Thus, the discrepancies between the calculated and measured value of $\hat{\alpha}^{49}$ will increase from 4.5 percent to ~ 6.7 percent. Correspondingly, the calculated value of $\hat{\alpha}^{41}$ is going to decrease by ~ 3.7 percent and be in better agreement with the value derived from the burnup data. The calculated ratio $\hat{\sigma}_a^{41}/\hat{\sigma}_a^{49}$ will decrease ~ 1.5 percent.

TABLE VII

SUMMARY OF THE CROSS SECTION DATA FOR Pu ISOTOPES

Resonance Integral, I_{∞} for Infinite Dilution

I_{∞} for a 0.414 eV Cutoff (barns)					2200 m/sec Values (barns)			
Isotope	Fission		Capture		Isotope	Cross Section	BNWML	ENDF/B-II
	BNWML	ENDF/B-II	BNWML	ENDF/B-II				
^{239}Pu	373	368	209	215	^{239}Pu	σ_a	1008.1	1012.9
^{240}Pu			8502	8519		σ_f	742.4	741.6
^{241}Pu	532	599	174	125		σ_c	265.7	271.3
^{242}Pu			1113	1130		ν	2.871	2.8799
						α	0.3580	0.3659
						η	2.114	2.1085
					^{241}Pu	σ_a	1391	1375.4
						σ_f	1009	1007.3
						σ_c	382	368.1
						ν	2.969	2.934
						α	0.379	0.3654
						η	2.154	2.149
					^{240}Pu	σ_c	282	290
					^{242}Pu	σ_c	30.0	30.0

The ~ 3 percent difference in the 2200 m/sec capture cross section shown in Table VIII, for ^{240}Pu will propagate to an ~ 1 percent change in the calculated effective value. The current best estimate of the 2200 m/sec cross section for ^{242}Pu is near 18.5 b rather than the 30 b value and this new value will be in Version III of ENDF/B. Thus, the calculated ratio $\hat{\sigma}_a^{42}/\hat{\sigma}_a^{49}$ is expected to decrease significantly. The shape of the cross section for ^{239}Pu between the BNWML and ENDF/B-II are negligible; therefore, no change is expected⁽⁵⁸⁾ in the ratios considering this as a source.

The resonance integral differences shown in Table VIII are not expected to cause the conclusions of the comparisons to change. The only cross sections which are expected to affect the calculated ratios are the ^{240}Pu and ^{242}Pu cross sections because the nonthermal absorption rates in the fissile isotopes are negligible. However, the differences in the ^{240}Pu and ^{242}Pu resonance integral values are small.

Thus, very little change (less than a few percent) would occur in the results presented in Table IV by substituting the improved versions of the codes (HRG-3 and BRT-I) and the use of Version-II ENDF/B cross sections. The changes are in some cases a reduction in the discrepancies noted (e.g., $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$ and $\hat{\alpha}^{41}$) and in other cases increase the discrepancy (e.g., $\hat{\alpha}^{49}$). Although the discrepancy between the calculated and measured values of $\hat{\alpha}^{49}$ is < 2 standard deviations, the thermalization calculation and/or the thermal neutron cross sections for ^{239}Pu might be judged inaccurate and causing the noted discrepancy. Clearly, a homogeneous representation of the 19-rod cluster is inadequate for calculating the nonthermal absorption rates in ^{240}Pu and ^{242}Pu and is the major cause of the discrepancy in the ratio $\hat{\sigma}_a^{40}/\hat{\sigma}_a^{49}$.

SUMMARY AND CONCLUSIONS

Two methods for deducing ratios of effective cross sections from isotopic data have been presented. One method uses equations which are explicit functions of the concentrations and this method yields ratios which are a function of exposure. The other method uses equations which are implicit functions of the concentrations and this method yields ratios which are constant or have a predetermined functional relationship with respect to exposure. Where applicable the preferred method is the latter or integrated equation approach. It provides a more rigorous statistical analysis of the data and requires no a priori values to be assumed for the cross section ratios. In this method, all the concentration data can be fit simultaneously and the method correctly accounts for correlated uncertainties. Both methods utilize specially developed programs of least squares technique to fit the transmutation relationships to the isotopic data. The coefficients of these fits are the ratios of effective cross sections.

Measurements of fuel isotopic composition have been obtained as functions of exposure from fuels irradiated in various power reactors. These data have been fitted using these methods to obtain ratios of effective cross sections. The results of the fits obtained from Al-Pu fuels irradiated in PRTR have been shown to illustrate the application of the technique. These data were fit with good statistical precision basically because the conditions under which these fuels were irradiated were well known and free from extraneous perturbations to the neutron spectrum such as those resulting from control rod effects.

Calculations were made to obtain results for comparison with the value deduced from the experimental data. In order to bring the calculated and deduced values of $\hat{\alpha}^{49}$ in agreement would require about a 4-5 percent decrease in the 2200 m/sec value of $\hat{\alpha}$ for ^{239}Pu from the value used in our analysis. Correspondingly, the current value on ENDF/B-II would have to be decreased by ~ 6 percent to force agreement. A more likely candidate in the cross sections themselves of this discrepancy is the variation of $^{239}\alpha$ with energy in the thermal neutron energy range. However, the status of knowledge on the thermal cross sections for ^{239}Pu has improved over recent years to the point where the uncertainties associated with these data are not large enough to account for the discrepancy. ⁽⁵⁸⁾ Therefore, the more logical candidate for the cause of this discrepancy is inadequacies in the THERMOS code. The results of other plutonium fueled reactor studies ^(59,60) support this contention.

The availability of data on the ratios of effective cross sections are important for testing the accuracy of burnup calculations. Where these ratios can be derived with good statistical precision, they are a sensitive test of the calculations and can be used as the criteria for judging the accuracy of unit cell calculations, as shown by the example presented in this paper. For those data, where the ratios are less certain due to spectral perturbations during burnup, they cannot be used singly as a criteria for testing the calculations. ⁽¹⁵⁾ Nevertheless, these ratios are still a useful adjunct to the isotopic data in theory-experiment correlations in that they can point to gross inadequacies in the calculations.

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FOOTNOTES

- (a) Now the current best value for the Cs^{137} half life appears to be 30.12 ± 0.21 yrs. reported by S. A. Reynolds and E. I. Wyatt in the "Analytical Chemistry Division Annual Progress Report for Period Ending October 31, 1966," ORNL-4039 (January 1967).
- (b) This fitting process involves the more familiar least squares method and is accomplished using Program Learn. The analysis logic of Program Learn is a special case of Program Likely. It is obtained by setting the span of the vector fields $\underline{q}$ and $\underline{y}$ to one (i.e., $J = L = 1$) and particularizing the implicit vector theory $\underline{q}(\underline{y}, \underline{p}) = 0$ to the explicit scalar theory $y = y(A)$.
- (c) A recent report: M. G. Cabell and M. Wilkins, "An Isometric State of ^{241}Pu ," J. Inorg. Nucl. Chem., 33, 903 (1971), gives a 15.16 year half life for ^{241}Pu .

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