BNWL-1523
Special
Distribution

CALCULATION OF ^{236}Pu AND ^{238}Pu
IN THE DISCHARGE FUEL
OF LIGHT WATER POWER REACTORS

D. L. Prezbindowski

September 1970

AEC RESEARCH &
DEVELOPMENT REPORT

BNWL-1523

NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

PACIFIC NORTHWEST LABORATORY

RICHLAND, WASHINGTON

operated by

BATTELLE MEMORIAL INSTITUTE

for the

UNITED STATES ATOMIC ENERGY COMMISSION UNDER CONTRACT AT(45-1)-1830

3 3679 00061 7557

BNWL-1523
Special Distribution

CALCULATION OF ^{236}Pu AND ^{238}Pu
IN THE DISCHARGE FUEL OF LIGHT WATER POWER REACTORS

D. L. Prezbindowski

Analytical Physics Section
Reactor Physics Department

September 1970

BATTELLE MEMORIAL INSTITUTE
PACIFIC NORTHWEST LABORATORIES
RICHLAND, WASHINGTON 99352

CALCULATION OF ^{236}Pu AND ^{238}Pu
IN THE DISCHARGE FUEL OF LIGHT WATER POWER REACTORS

D. L. Prezbindowski

ABSTRACT

An important consideration in the design and operation of reactor fuel fabrication plants handling plutonium enriched fuel is the isotopic composition of the plutonium that the plant will receive. The plutonium isotopes produced in light water reactors emit a variety of alpha and gamma radiations and neutrons from spontaneous fission. The exact magnitude and character of radiation being produced by the plutonium enriched fuel being fabricated dictates the amount and kind of shielding required for the fuel fabrication process. A study has been performed to determine the isotopic composition of discharged fuel produced by LWR's. Particular emphasis was placed on the determination of ^{236}Pu and ^{238}Pu concentrations since data on these isotopes are not as plentiful as data on the other plutonium isotopes.

TABLE OF CONTENTS

	<u>Page</u>
ABSTRACT	iii
LIST OF TABLES	v
LIST OF FIGURES	vi
INTRODUCTION	1
SUMMARY OF RESULTS AND CONCLUSIONS	3
THE REACTOR MODEL	4
CALCULATIONAL METHODS AND ASSUMPTIONS	9
Description of ZODIAC	9
Simplified Decay Scheme	10
Reactor Representation	10
Cross Section Data (Hybrid).	12
Cross Section Data (1969 Evaluation)	15
RESULTS AND CONCLUSIONS	15
Yankee Rowe PWR	15
Fort Calhoun PWR	16
BOILING WATER REACTOR	23
RECOMMENDATIONS FOR FUTURE WORK	24
ACKNOWLEDGEMENTS	55
APPENDIX I	I-1

LIST OF TABLES

<u>Number</u>	<u>Title</u>	<u>Page</u>
I	Summary Table on PWRs	6
II	Summary Table on BWRs	7
III	Reactor Characteristics-Average	8
IV	Actual and Simplified Transmutation Path used in the ZODIAC Program	10a
V	Simplified ZODIAC Transmutation and Decay Scheme . .	11
VI	Yankee Rowe Schedule	13
VII	FORT CALHOUN Schedule	13
VIII	Yankee Isotopic Concentrations	14
IX	FORT CALHOUN Isotopic Concentrations	14
X	Cosine Axial Exposure Weighted at 17055 MWD/MTM Avg. Element Exposure	19
XI	Cosine Axial Exposure Weighted at 28000 MWD/MTM Avg. Element Exposure	20
XII	Multiregion ZODIAC at 28000 MWD/MTM Avg. Element Exposure	21

LIST OF FIGURES

<u>Number</u>	<u>Title</u>	<u>Page No.</u>
1	ZODIAC Calculated ^{239}Pu - ^{242}Pu Isotopic Concentration vs Analytical Data	27
2	ZODIAC Calculated ^{236}Pu Isotopic Concentration vs Analytical Data	28
3	ZODIAC Calculated ^{236}U , ^{237}Np , ^{238}Pu Isotopic Concentration vs Analytical Data	29
4	FORT CALHOUN - Dissolved Boron Concentration Evaluation as a Function of Burnup	30
5	Plot of Burnup for FORT CALHOUN Versus	31
6	Plot of ^{236}Pu Versus ^{235}U for FORT CALHOUN	32
7	Plot of ^{238}Pu Versus ^{235}U for FORT CALHOUN	33
8	Plot of ^{239}Pu Versus ^{235}U for FORT CALHOUN	34
9	Plot of ^{240}Pu Versus ^{235}U for FORT CALHOUN	35
10	Plot of ^{241}Pu Versus ^{235}U for FORT CALHOUN	36
11	Plot of ^{242}Pu Versus ^{235}U for FORT CALHOUN	37
12	Plot of ^{237}Np Versus ^{235}U for FORT CALHOUN	38
13	Regional Power Versus Axial Position for a FORT CALHOUN Fuel Element	39
14	Regional Exposure Versus Axial Position for a FORT CALHOUN Fuel Element	40
15	FORT CALHOUN - PWR Average Exposure Versus Position Along Rod	41
16	FORT CALHOUN - PWR ^{236}Pu Concentration Versus Position Along Rod at 28,000 MWD/MTM Average.	42
17	FORT CALHOUN - PWR ^{238}Pu Concentration Versus Position Along Rod at 28,000 MWD/MTM Average	43
18	FORT CALHOUN - PWR ^{239}Pu Concentration Versus Position Along Rod at 28,000 MWD/MTM Average	44

LIST OF FIGURES (Continued)

<u>Number</u>	<u>Title</u>	<u>Page No.</u>
19	FORT CALHOUN - PWR ^{240}Pu Concentration Versus Position Along Rod at 28,000 MWD/MTM Average	45
20	FORT CALHOUN - PWR ^{241}Pu Concentration Versus Position Along Rod at 28,000 MWD/MTM Average	46
21	FORT CALHOUN - PWR ^{242}Pu Concentration Versus Position Along Rod at 28,000 MWD/MTM Average	47
22	FORT CALHOUN - PWR Exposure Versus Position Along Rod	48
23	FORT CALHOUN - PWR ^{236}Pu Concentration Versus Position Along Rod at 17055 MWD/MTM Average	49
24	FORT CALHOUN - PWR ^{238}Pu Concentration Versus Position Along Rod at 17055 MWD/MTM Average	50
25	FORT CALHOUN - PWR ^{239}Pu Concentration Versus Position Along Rod at 17055 MWD/MTM Average	51
26	FORT CALHOUN - PWR ^{240}Pu Concentration Versus Position Along Rod at 17055 MWD/MTM Average	52
27	FORT CALHOUN - PWR ^{241}Pu Concentration Versus Position Along Rod at 17055 MWD/MTM Average	53
28	FORT CALHOUN - PWR ^{242}Pu Concentration Versus Position Along Rod at 17055 MWD/MTM Average	54

INTRODUCTION

Purpose

Additional reactor fuel fabrication capability must be designed and built to accomodate the growing demand for reactor fuel. In order to properly design the radiation shielding for these facilities it is necessary to determine the strength of the important radiation sources in the plutonium enriched fuel. The amount and kind of radiation that is produced by plutonium enriched fuels for fast and thermal reactors in a fuel fabrication plant may have a profound influence upon fuel fabrication methodology and cost. This influence is reflected in the fuel cycle economics of the reactors for which the fuel is being fabricated. Two sources of alpha (α) and gamma (γ) radiation in the fuel which must be considered are the plutonium isotopes ^{236}Pu and ^{238}Pu . These isotopes are not highly important to the determination of reactivity and power distribution in power reactors and are not routinely included in reactor burnup calculations as are the ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu isotopes.

Because of the importance of an accurate estimation of the ^{236}Pu and ^{238}Pu concentrations to shielding design, the concentrations of these isotopes were calculated in the discharge fuel of typical PWR power reactors. The results of these calculations establish a firm foundation for the source definition of shielding calculations and help to verify the accuracy and determine the usefulness of other studies in this area. The parametric studies performed in Reference 1 provide

isotopic composition versus a variety of reactor parameters. These results are very useful for predicting ^{236}Pu and ^{238}Pu concentrations of typical reactors if the results can be proven to be sufficiently accurate. The ALTHAEA computer code used in the study must be normalized to experimental results or to a more accurate analytical method. The results given in Reference 1 were based on a normalization to the computer code ZODIAC,⁽²⁾ a 1-dimensional burnup code. However, cross sections and code improvements have been made in ZODIAC since the initial calibration which may have made the calibration obsolete. Because of the requirement of a high degree of accuracy and reliability in the determination of the ^{236}Pu and ^{238}Pu concentrations, a program of ZODIAC calculations on two PWR plants was established to verify the following quantities specified in Reference 1:

- a. Exposure
- b. Isotopic concentration values for ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu , for the Yankee Rowe and Fort Calhoun Reactors.

Care has been taken to examine the effects of the axial spatial averaging of the isotopic concentrations in a PWR fuel element. Average concentrations have been calculated for two different axial flux shape models: a cosine flux shape throughout the irradiation and an irradiation-dependent axial flux shape.

SUMMARY OF RESULTS AND CONCLUSIONS

A compilation of pounds of UO_2 discharged annually for PWRs and BWRs and reactor design characteristics which are important to ^{236}Pu and ^{238}Pu production was made from available Preliminary and Final Safety Analysis Reports (PSARs and FSARs). Average PWR and BWR design characteristics were defined from this information. The strongest ^{236}Pu and ^{238}Pu producer of all PWRs in the reactor list was determined to be Yankee Rowe from the ALTHAEA results.⁽¹⁾

ZODIAC calculated isotopic concentrations were correlated with Yankee Rowe experimental data⁽²⁾ and were found to be within the experimental error of the data when the hybrid cross section library is used. Isotopic concentrations were calculated for the Fort Calhoun reactor, which is representative of the average modern PWR, in the plutonium production sensitive design characteristics of moderator to fuel ratio, enrichment, and fuel diameter. Comparison of ZODIAC and ALTHAEA results show that ALTHAEA underpredicts plutonium isotopic concentrations at high exposures by as much as 10-20% but predicts basic isotopic trends adequately. Exposures calculated with ZODIAC and ALTHAEA were in agreement and ZODIAC was in good agreement with Yankee Rowe experimental results. ZODIAC calculations with and without boron in Fort Calhoun have shown that the influence of boron on the isotopic concentrations is of the order of a few percent.

An axial average of ^{236}Pu and ^{238}Pu concentration was performed for the Fort Calhoun fuel at 17055 MWD/MTM,^(*) average fuel exposure. The average was based on an assumed cosine axial flux shape throughout the irradiation. These averages were 22.3% higher for ^{236}Pu concentration and 37.6% higher for ^{238}Pu concentration than the concentrations obtained

^(*) Megawatt days/metric ton of uranium and plutonium metal present before irradiation.

from the ZODIAC isotopic values at 17055 MWD/MTM. This example illustrates the importance of spatial averaging in determining fuel average isotopic concentration.

An axial average of ^{236}Pu and ^{238}Pu isotopic concentration assuming a cosine flux shape was also performed at 28000 MWD/MTM. The calculated isotopic concentrations are 8.793×10^{-2} WPPM (weight parts per million) and 1.548×10^4 WPPM for ^{236}Pu and ^{238}Pu , respectively. These concentrations are 17% and 29% above the non-spatial-average values: A more realistic multi-region ZODIAC burnup calculation gives isotopic concentrations 6% and 4% above the non-average values. This comparison indicates the degree of overestimation associated with the cosine shape averaged values.

Because of the complex spatial shape of flux and moderator-to-fuel ratio in a BWR it is necessary to do a three-dimensional analysis of the BWR in order to obtain appropriate average isotopic compositions. Such an analysis was not performed in the study reported here, but some discussion of the problem is given in a later section.

THE REACTOR MODEL

The first step in defining the amount of ^{236}Pu and ^{238}Pu available in discharged reactor fuel is to describe the reactors from which the fuel is obtained. The reactor description used in this study focused on all characteristics which influence the ^{236}Pu and ^{238}Pu content, but

especially on the important characteristics of fuel diameter, moderator-to-fuel ratio and average fuel enrichment.

A list of PWRs and BWRs was compiled from various sources in the literature. The data for these reactors was compiled from preliminary and final safety analysis reports. Table I and Table II list design characteristics for the PWRs and BWRs, respectively. Table III lists average design characteristics for the PWRs and BWRs in Tables I and II. The data on Tables I through III reveal the average and extremes of variation of the important reactor design characteristics. The gaps in the tables are due to the unavailability of safety analysis reports for the reactors involved.

Having identified as far as possible the characteristics for PWRs and BWRs which will be operating in the immediate future, the next step is to relate the reactor design characteristics which are most important in their effect on the ^{236}Pu and ^{238}Pu concentration in the fuel. An examination of Appendix B of Reference 1 reveals the following trends relating reactor design characteristics of BWRs and PWRs to the production of ^{238}Pu and ^{237}Np , the precursor of ^{236}Pu . The trends listed are in order of importance.

1. Moderator-to-fuel ratio:

- inversely related to ^{237}Np production
- inversely related to ^{238}Pu production

2. Fuel enrichment:

- directly related to ^{237}Np production
- inversely related to ^{238}Pu production

TABLE I. SUMMARY TABLE ON PWRs

Reactor	Fuel Diameter (in)	Moderator/ Fuel Ratio	Enrichment	
			Outer Region Initial Core	Equilibrium
Beaver Valley	0.3669	1.67	3.2	3.2
Calvert Cliffs	0.382		3.03	
Connecticut Yankee	0.3835	1.533	3.67	
Crystal River-3	0.362	1.68	2.90	2.94
Donald Cook				
Diablo Canyon-1	0.3669	1.67	2.67	
Diablo Canyon-2	0.3669	1.67	3.70	
Fort Calhoun	0.383	1.60	2.95	3.05
Indian Point-2	0.3669	1.67	2.68	2.92
Indian Point-3	0.3669	1.67	3.20	
Kewaunee	0.3669	1.60	3.15	3.38
Main Yankee	0.382	1.61	3.01	
Midland-1	0.370	1.65	2.64	
Oconee-1	0.362	1.79	2.77	
Oconee-2	0.362	1.68		2.47
Oconee-3				
Oyster Creek-2	0.362	1.68	2.90	2.94
Palisades	0.359	1.70	2.74	2.74
Point Beach-1	0.3669	1.60	2.80	
Point Beach-2	0.3669	1.60	2.80	3.05
Prarie Island-1	0.3669	1.60	3.15	3.38
Prarie Island-2	0.3669	1.60	3.15	3.38
Robinson-2	0.3669	1.675	2.73	
Russellville	0.362	1.68	2.90	2.94
Salem-1	0.3669	1.67	3.20	
Sequoia-1	0.3659	1.68	3.20	
Sequoia-2				
Surry-1	0.3669	1.675	2.94	3.58
Surry-2	0.3669	1.675	2.94	3.58
Three Mile Island	0.362	1.67	2.90	2.94
Turkey Point-3	0.3669	1.67	2.73	
Turkey Point-4	0.3669	1.67	2.73	
Zion-1	0.3669	1.72	3.20	
Zion-2	0.3669	1.72	3.20	
Yankee Rowe	0.286	1.286	3.4%	
Core I & II				
Average	0.3678	1.659	2.968	3.099

TABLE II. SUMMARY TABLE ON BWRs

<u>Reactor</u>	<u>Fuel Diameter (in)</u>	<u>Moderator/ Fuel Ratio</u>	<u>Average Enrichment Initial Core</u>
Arnold	0.487	1.587	2.23
Bell	0.488	1.586	2.23
Big Rock Point			
Brown's Ferry-1		1.548	2.19
Brown's Ferry-2			
Brown's Ferry-3		1.58	2.19
Brunswick 1	0.487		2.25
Cooper	0.488	1.578	2.15
Dresden-1			
Dresden-2	0.488	1.47	2.36
Dresden-3	0.488	1.47	2.36
Fitzpatrick	0.487	1.587	2.23
Hatch			
LaCrosse	0.350	2.038	3.63
Millstone Point	0.488	1.47	2.07
Monticello	0.488	1.55	2.03
Nine Mile Point	0.488	1.47	2.10
Oyster Creek-1	0.488	1.40	2.10
Pilgrim	0.488	1.58	2.18
Quad Cities I & II	0.488	1.586	2.18
Shoreham	0.488	1.586	2.28
Vermont Yankee	0.488	1.58	2.29
Peach Bottom-1	0.488	1.586	2.19
Peach Bottom-2	0.488	1.586	2.19
Peach Bottom-3	<u>0.488</u>	<u>1.586</u>	<u>2.19</u>
Average	0.4805	1.580	2.263

TABLE III. REACTOR CHARACTERISTICS - AVERAGE

PWR (Averaged using 34 reactors)

Fuel diameter (pellet)	0.3677 in.
Moderator/fuel ratio	1.659
Enrichment (Region 3)	2.975
Power	2347 MWth
Average Discharge Exposure, 1st core	17437 MWD/MTM
Average Discharge Exposure, Equilibrium	29539 MWD/MTM
Lattice Pitch	0.544 in.
Core diameter	115.66 in.
Bundle Arrangement	15 x 15 or 14 x 14

BWR (Averaged using 20 reactors)

Fuel diameter (pellet)	0.4805 in.
Moderator/fuel ratio	1.580
Enrichment	2.263
Power	2271.6 MWth
Average Discharge Exposure	19609 MWD/MTM
Lattice pitch	0.373 in.
Core diameter	157.87 in.
Bundle Arrangement	7 x 7

3. Fuel diameter:
 inversely related to ^{237}Np production
 directly related to ^{238}Pu production
4. Stainless steel clad yields slightly higher values than
 zirconium for ^{237}Np and ^{238}Pu for both BWRs and PWRs.

Scanning Table I with the four trends in mind it can be seen that Yankee Rowe will produce more ^{238}Pu and ^{236}Pu than any other PWR listed primarily because of its combination of high enrichment and low moderator-to-fuel ratio.

From Table I and Table III it can be seen that the Fort Calhoun reactor has a slightly lower equilibrium core enrichment: 3.05 vs 3.099 for the PWR average, slightly lower moderator-to-fuel ratio: 1.60 vs 1.659 for the PWR average and a higher fuel diameter: 383 vs 3678 for the PWR average.

The lower Fort Calhoun moderator-to-fuel ratio tends to increase Pu production. The lower Fort Calhoun core enrichment and higher fuel diameter tend to increase ^{238}Pu production and decrease ^{236}Pu production. As a result, Fort Calhoun is considered representative of the average PWR with regard to the reactor characteristics used to calculate plutonium production. It is conservative in that it tends to have a slightly higher Pu production rate than a reactor with the average characteristics. Fort Calhoun was chosen as a representative PWR based on characteristics that influence the Pu production rate per unit weight of fuel. No attempt was made to match other reactor characteristics which do not influence the Pu production rate per unit weight of fuel.

CALCULATIONAL METHODS AND ASSUMPTIONS

DESCRIPTION OF ZODIAC

The ZODIAC code has gone through a succession of evolutionary developments as documented in References 4 and 5. Recent significant improvements to the code include the implementation of the intermediate resonance formulation in the HRG portion of ZODIAC, and a new evaluation of much of the cross-section data.

The ZODIAC code is designed to do a multigroup, multiregion, burnup analysis of nuclear reactor fuel. ZODIAC is a combination of two operating systems of reactor physics codes which are chained together for

efficient use. The first system known as the HTH code,⁽⁶⁾ is composed of the reactor physics codes HRG,⁽⁷⁾ THERMOS,⁽⁸⁾ and HFN.⁽⁹⁾ The second system is a burnup package consisting of CLERK,⁽¹⁰⁾ and ALCHEMY.⁽¹¹⁾

In performing a reactor burnup analysis the multigroup cross sections are determined by the codes HRG and THERMOS, the multigroup fluxes and the reactivity are determined by HFN, and the isotopic transmutations are determined by ALCHEMY. The multiregion burnup is limited to one space dimension.

Simplified Decay Scheme

A detailed decay and transmutation scheme which accounts for the important contributions to ^{236}Pu and ^{238}Pu is given in Table IV. Space limitations in the ZODIAC program preclude the representation of this decay scheme in its full generality. A simplified transmutation and decay scheme which was used in the program is given in Table IV and V. The validity of the simplifying assumptions made in defining the simplified scheme is evident from the good correlation of ZODIAC and Yankee Rowe burnup data as shown in Figure 1.

Reactor Representation

The isotopic compositions calculated by ZODIAC for Yankee Rowe and Fort Calhoun are representative of the fuel rods located at least several rows away from the fuel bundle edge in these cores (i.e., the asymptotic zone). The fuel temperature used in the Yankee Rowe calculation (708°C) is representative of the temperature of the experimental fuel samples to which it is compared. The temperature used in the Fort Calhoun calculation is the core average (871°C).

The ZODIAC isotopic calculations are based on a fuel cell model of a fuel pin in surrounding water for the HRG and THERMOS portion of the calculation. The HFN model consists of a single fully reflected

TABLE IV. ACTUAL AND SIMPLIFIED TRANSMUTATION PATHS USED IN THE ZODIAC PROGRAM

TABLE V

SIMPLIFIED ZODIAC TRANSMUTATION AND DECAY SCHEME*

*Removal of an isotope by fission and capture processes other than those represented here was also accounted for.

homogenized fuel region. This model was chosen since the calculation of k_{eff} and reactor power distributions was not required. The neutron spectrum cross section averaging schedule is given in Tables VI and VII for the ZODIAC calculations of Yankee Rowe and Fort Calhoun, respectively. The fuel cell homogenized isotopic compositions used in the ZODIAC calculation are given in Tables VIII and IX for Yankee Rowe and Fort Calhoun, respectively.

Cross Section Data (Hybrid)

One of the fundamental problems in performing reactor calculations is to decide what cross section data to use. The Battelle Northwest Master Library (BNWML) represents a cross section data file which has been compiled over a period of years. The data file is periodically updated upon recent evaluations and experimental-analytical correlations are then made to determine the effect of the updates on reactor calculations. The Yankee Rowe Cores I and II have been used as a basis for many of these correlations.

Comparisons were made of ZODIAC calculations on Yankee Rowe using the 1969 evaluation version of the BNWML and previous calculations made using the 1965 version⁽¹²⁾ of the BNWML. The comparisons showed that the 1965 data calculations had a much better theory-experiment correlation of ^{239}Pu than the calculations using 1969 data. The calculations using the 1969 data in which ^{238}U has an infinite dilute resonance integral of 269 barns produced ^{239}Pu isotopic concentrations which were 12% lower than the fitted experimental data. Calculations using ^{238}U cross sections with an infinite dilute resonance integral of 282 barns (1965 evaluation) gave a good correlation of theory and experiment.

TABLE VI. HRG-THERMOS AND HFN FLUX WEIGHTING
SCHEDULE FOR EFFECTIVE CROSS SECTIONS
FOR THE YANKEE ROWE REACTOR CALCULATION

<u>HRG-THERMOS</u>	<u>HFN</u>
0*	60*
40	80
100	133
200	166
400	250
600	300
800	350
	450
	500
	550
	666
	733
	900

TABLE VII. HRG-THERMOS AND HFN FLUX WEIGHTING
SCHEDULE FOR EFFECTIVE CROSS SECTIONS
FOR THE FORT CALHOUN REACTOR CALCULATION

<u>HRG-THERMOS</u>	<u>HFN</u>
0*	60*
40	80
100	133
200	166
400	250
600	300
800	350
1100	450
1300	500
1500	550
	666
	733
	900
	1000
	1200
	1400
	1600

*Full Power Days at 28.2 KW/kg

TABLE VIII. PIN CELL NUMBER DENSITIES FOR
THE YANKEE ROWE REACTOR

*	Fuel	Clad	Moderator
H			5.2313 -02
O	4.41315 -02		2.6156 -02
Fe		5.593 -02	
Cr		1.598 -02	
Ni		7.991 -03	
U-238	2.1302 -02		
U-235	7.5933 -04		
U-236	4.4 -06		

TABLE IX. PIN CELL NUMBER DENSITIES FOR THE
FORT CALHOUN REACTOR

*	Fuel	Clad	Moderator
H			4.85176 -02
O	4.42517 -02		2.42588 -02
Zr		4.06582 -02	
U-235	6.74839 -04		
U-238	2.14510 -02		

*Units in Atoms/barn-cm.

In order to combine the best features of both data sets, a hybrid data file was created in which the 1965 data was used for ^{238}U and the 1969 data was used for the remaining isotopes. Using the hybrid data set, a ZODIAC calculation was performed on Yankee Rowe Cores I and II. A comparison of the results obtained using the hybrid data set with results obtained using the 1969 data set is given in Figure 1, where the correction of the previous underestimation of ^{239}Pu can be seen.

Cross Section Data (1969 Evaluation)

The cross section evaluation performed by B. R. Leonard, Jr. for eight of the isotopes used in the ZODIAC calculations is described in Appendix I. The data resulting from this evaluation are included in the 1969 data file.

RESULTS AND CONCLUSIONS

Yankee Rowe PWR

The correlation of isotopic concentrations calculated with ZODIAC and experimental concentrations from Yankee Rowe is quite important because Yankee Rowe is the strongest producer of ^{236}Pu and ^{238}Pu on the PWR reactor list and because it demonstrates the accuracy of ZODIAC in the calculation of isotopic compositions for PWRs.

Figure 1 contains a comparison of Yankee Rowe isotopic data fit with the least squares fitting program DBUFIT⁽¹³⁾ and isotopic concentrations calculated with ZODIAC using both the hybrid and the 1969 evaluation cross section data set. The ZODIAC results using the 1969 evaluation cross section data show significant discrepancies, especially in the correlation of the ^{239}Pu isotopic concentration, where

the calculated results are 12% lower than the fitted curve at 12000 MWD/MTM. The ZODIAC results obtained using a combination of the 1969 evaluation cross section data and the ^{238}U cross section data from the previous evaluation (hybrid cross section data set) are within the scatter of the point data everywhere.

Figure 2 gives the fitted Yankee Rowe ^{236}Pu data and ^{238}Pu concentrations calculated with ZODIAC. It can be seen that the ^{236}Pu concentration for both the hybrid and the 1969 evaluation cross section data is generally lower than the experimental fitted curve - about 8% lower at the highest exposures; however, the calculated points lie within the uncertainty bars attached to the point data everywhere.

Figure 3 gives the fitted Yankee Rowe ^{236}U , ^{237}Np and ^{238}Pu data and the ^{236}U , ^{237}Np and ^{238}Pu concentrations calculated with ZODIAC. It can be seen that the correlation of the ^{237}Np data is quite good for both the hybrid and the 1969 evaluation data sets. The calculated results tend to be a little lower than experiment, especially at mid and lower exposures. This may contribute to the lower calculated values of ^{238}Pu and ^{236}Pu since ^{237}Np is a precursor of both isotopes. The ^{238}Pu correlation with the experimental data is satisfactory throughout; the calculated points fall within the uncertainty of the experimental data everywhere.

Fort Calhoun PWR

The Fort Calhoun PWR has design characteristics which are close to the average characteristics of interest to plutonium isotopic production for the reactors in the PWR reactor list. The isotopic concentrations

produced by this reactor should be representative of the average future PWR. Considerable time was spent in determining the sensitivity of the isotopic concentrations to the amount of dissolved boron in the moderator. The amount of dissolved boron in the moderator throughout reactor operation is not published in the FSAR but must be calculated. The calculated boron concentration in the moderator of Fort Calhoun during one year of operation under equilibrium core conditions is given in Figure 4 as calculated with ZODIAC. Figures 5 through 12 give isotopic concentrations and burnup calculated by ZODIAC with and without boron and also ALTHAEA results for ^{238}Pu , ^{239}Pu , ^{240}Pu and ^{237}Np . The differences between the isotopic concentrations calculated with and without boron are not large for any isotope and for this reason the calculations with boron were not carried to full exposure.

The ALTHAEA results were obtained by interpolation from the tables in Reference 1. It can be seen that the ALTHAEA results for ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu are generally low at high exposures compared with ZODIAC results (i.e., ^{239}Pu is 5% low, ^{240}Pu is 2% low, ^{241}Pu is 8% low, and ^{242}Pu is 20% low). In addition, ^{237}Np is 3% low, and ^{238}Pu is 8% high at some low exposures and 11% low at high exposures. This shows that ALTHAEA results for plutonium isotopes, except ^{240}Pu at high exposure, are likely to be significantly under predicted. The ^{236}Pu information calculated with ALTHAEA which has been published is limited to a value for one PWR at one exposure, so that one must rely on ZODIAC results for establishing ^{236}Pu concentrations.

A glance at curves of ^{236}Pu and ^{238}Pu for either Fort Calhoun or Yankee Rowe shows that the slopes of the curves are quite steep at higher

exposures. This means that errors in the calculation of exposure by a code could cause significant errors in the ^{236}Pu , ^{238}Pu determination. Figure 5 contains a comparison of exposure vs ^{235}U for Fort Calhoun as calculated by ZODIAC and ALTHAEA. It can be seen that ZODIAC and ALTHAEA are in good agreement with regard to the exposure calculation. Figure 1 shows that the ZODIAC calculation of exposure is in good agreement with the experimental determination of exposure for Yankee Rowe.

Having determined the variation of ^{236}Pu and ^{238}Pu with exposure at a given point in Fort Calhoun, attention must be turned to the determination of appropriate averages of these isotopes in a fuel element. The major variation in exposure in a fuel element is along the Z axis. Because of the curvature in the variation of ^{236}Pu and ^{238}Pu with exposure, one must calculate the average ^{236}Pu and ^{238}Pu present in an element from the curve of ^{236}Pu and ^{238}Pu vs axial position rather than simply using the average element exposure and the plot of ^{236}Pu and ^{238}Pu vs exposure.

This averaging procedure was performed for Fort Calhoun at 17055 MWD/MIM and 28,000 MWD/MIM average element exposure. The process is represented in Figures 15 through 28. The exposure curves in Figures 15 and 22 were obtained by assuming a cosine shape for the axial flux. These curves were used in conjunction with the curves of isotopic composition vs exposure to obtain plots of isotopic composition vs axial position. The curves of isotopic composition vs position were integrated to determine the axial average isotopic composition. The average ^{236}Pu and ^{238}Pu concentrations obtained by this process is given in Tables X and XI. At 17055 MWD/MIM these averages are 22% and 37%

TABLE X

FORT CALHOUN AVERAGE ISOTOPIC CONCENTRATION - COSINE AXIAL
EXPOSURE WEIGHTED AT 17055 MWD/MTM AVERAGE ELEMENT EXPOSURE

$\frac{n}{b-cm}^*$		$A \text{ ppm}^{**}$	$W \text{ ppm}^{***}$
$^{239}\text{Pu} = 1.07046 \times 10^{-4}$	^{239}Pu	6.8914×10^5	6.8786×10^5
$^{240}\text{Pu} = 2.79795 \times 10^{-5}$	^{240}Pu	1.8013×10^5	1.8054×10^5
$^{241}\text{Pu} = 1.57231 \times 10^{-5}$	^{241}Pu	1.0122×10^5	1.0183×10^5
$^{242}\text{Pu} = 3.59385 \times 10^{-6}$	^{242}Pu	2.314×10^4	2.3384×10^4
$^{236}\text{Pu} = 6.5846 \times 10^{-12}$	^{236}Pu	4.239×10^{-2}	4.175×10^{-2}
$^{238}\text{Pu} = 9.9015 \times 10^{-7}$	^{238}Pu	6.37×10^3	6.336×10^3
<hr/>			
Total Pu = 1.553326×10^{-4}			

*nuclei/ 10^{-24} cm^3

**Atom parts per million

***Weight parts per million

TABLE XI

FORT CALHOUN AVERAGE ISOTOPIC CONCENTRATION - COSINE AXIAL
EXPOSURE WEIGHTED AT 28000 MWD/MTM AVERAGE ELEMENT EXPOSURE

$n/b\text{-cm}^*$	$A\text{ ppm}^{**}$	$W\text{ ppm}^{***}$
$^{239}\text{Pu} = 1.27136 \times 10^{-4}$	$^{239}\text{Pu} \quad 5.9646 \times 10^5$	5.9497×10^5
$^{240}\text{Pu} = 4.5887 \times 10^{-5}$	$^{240}\text{Pu} \quad 2.1530 \times 10^5$	2.1566×10^5
$^{241}\text{Pu} = 2.6443 \times 10^{-5}$	$^{241}\text{Pu} \quad 1.2407 \times 10^5$	1.2479×10^5
$^{242}\text{Pu} = 1.03569 \times 10^{-5}$	$^{242}\text{Pu} \quad 4.859 \times 10^4$	4.9082×10^4
$^{236}\text{Pu} = 1.90277 \times 10^{-11}$	$^{236}\text{Pu} \quad 8.929 \times 10^{-2}$	8.793×10^{-2}
$^{238}\text{Pu} = 3.32209 \times 10^{-6}$	$^{238}\text{Pu} \quad 1.559 \times 10^4$	1.543×10^4
<hr/>		
Total Pu = 2.13145×10^{-4}		

*nuclei/ 10^{-24} cm^3

**Atom parts per million

***Weight parts per million

TABLE XII

FORT CALHOUN AVERAGE ISOTOPIC CONCENTRATIONS CALCULATED BY MULTIREGION
ZODIAC AT 28000 MWD/MTM AVERAGE ELEMENT EXPOSURE

	<u>n/b-cm</u> [*]	<u>Wt ppm</u> ^{**}	<u>At ppm</u> ^{***}
²³⁶ Pu	1.71181-11	7.57011-2	7.68475-2
²³⁸ Pu	2.68015-6	1.19529+4	1.20320+4
²³⁹ Pu	1.36062-4	6.09351+5	6.10817+5
²⁴⁰ Pu	4.68562-5	2.10722+5	2.10349+5
²⁴¹ Pu	2.78874-5	1.25944+5	1.25194+5
²⁴² Pu	9.26835-6	4.20306+4	4.16080+4
Total Pu	222.75412-6		

*nuclei/10⁻²⁴ cm³

**Weight parts per million

***Atom parts per million

higher for ^{236}Pu and ^{238}Pu respectively than the values obtained using element average exposures and the ZODIAC isotopic curves. A similar situation exists at 28000 MWD/MTM where the spatial average values are 17.4% and 29.26% higher for ^{236}Pu and ^{238}Pu , respectively. However, it should be noted that the axial averaged values assuming a cosine power shape throughout the irradiation are actually overestimates of the ^{236}Pu and ^{238}Pu isotopic concentration. This is because the actual power shape flattens in the center as the irradiation proceeds.

In order to determine how much overestimation is built into the assumption of a cosine power shape, a multi-region axial burnup calculation was performed with ZODIAC. One-half the axial dimension of the Fort Calhoun fuel element was approximated by a 5 region slab with a reflection boundary on the left and a water stainless steel reflector region on the right. Figures 13 and 14 give the axial regional power and exposure, respectively. Information is given at three different times: 0 days, 301 days, and 997.7 days which correspond to 0, 8551, and 28342.3 MWD/MTM average exposure, respectively. The information at 301 days is presented to demonstrate how early in the exposure an essentially asymptotic power shape is achieved. Figure 13 shows the similarity between the power distribution at 301 and 997.7 days. Both distributions are in sharp contrast to the cosine shape at 0 days. Figure 14 gives the region exposure distributions at 301 and 997.7 days. Both distributions are flattened in the fuel center due to the flattened power shape. The isotopic concentrations for the plutonium isotopes determined by the multi-region ZODIAC calculation are given in Table XII. A comparison of Tables XI and XII indicates the differences between the

plutonium concentrations as calculated by the multi-region ZODIAC and assumed cosine shape methods. For ^{236}Pu and ^{238}Pu in particular the assumed cosine shape method increases ^{236}Pu and ^{238}Pu 17.4% and 29.56%, respectively, above the non-averaged values, while the multi-region ZODIAC calculation gives ^{236}Pu and ^{238}Pu concentrations which are respectively 6% and 4% above the non-averaged values at 28,000 MWD/MTM exposure. It is apparent that the cosine averaging method overpredicts ^{236}Pu and ^{238}Pu by 10-20% at these high exposures. These results indicate that the cosine flux shape assumption for axial averaging is not applicable for exposures greater than about 8551 MWD/MTM. At exposures greater than 8551 MWD/MTM the multi-region burn axial averaged values are close enough to the non-averaged values (less than 10% difference) so that averaging calculations may be unnecessary for some applications.

BOILING WATER REACTOR

There are important differences between the PWR and BWR which significantly affect the amount of ^{236}Pu and ^{238}Pu produced in the fuel. In the modern PWR, the reactivity is primarily controlled by dissolved boron which is uniformly distributed in the moderator. The reactor is designed to avoid significant boiling in the moderator so that the variation in the moderator-fuel ratio axially and radially is small. Because of dissolved boron control and variable fuel enrichment in the radial direction the radial peaking is kept to a minimum and the axial flux has a simple cosine shape at the beginning of exposure. These features make the calculation of element average concentrations for a PWR relatively straightforward.

In a BWR, however, there is a substantial variation in the moderator-to-fuel ratio in three dimensions. The flux shape axially and radially is dependent upon many other things, e.g., the control rod insertion pattern and the space dependent moderator-to-fuel ratio. For these reasons it is necessary to analyze the three-dimensional neutronic-thermo hydraulic characteristics and isotopic concentrations of a BWR in order to be able to calculate the average isotopic concentration in an element. There is no guarantee that the calculation of isotopic concentrations using a volume averaged moderator-to-fuel ratio and a volume average neutron flux will produce the proper element averages.

The FLARE⁽¹⁴⁾ code can provide three-dimensional analysis of the BWR for reasonable computer costs. It performs a three-dimensional neutronic-thermo hydraulic analysis of the BWR core and together with ISOCHECK⁽¹⁵⁾ produces a three-dimensional distribution of isotopic concentrations. The three-dimensional distribution may then be averaged over an element to give an element average. It is difficult to estimate whether the BWR or PWR produces more ²³⁶Pu and ²³⁸Pu on the average. Low local values of moderator-to-fuel ratio in the BWR point to high ²³⁶Pu, ²³⁸Pu production; however, low power levels in these same areas would tend to decrease the production. An estimate of the average BWR production awaits a three-dimensional treatment of the problem.

RECOMMENDATIONS FOR FUTURE WORK

In order to provide an adequate picture of the composition of plutonium fuels that will be passing through fuel fabrication plants, the following data should be generated:

1. Axial average plutonium, ^{236}Pu , ^{238}Pu and $^{239-242}\text{Pu}$ at given average discharge exposures for each and every operating PWR and BWR.
2. Volume average exposure of discharge fuel from all reactors in a specified group of reactors.
3. Volume average of the total plutonium production for any given reactor group per unit weight of fuel.
4. Total plutonium production from all reactors in a specified group.
5. Average plutonium isotope composition in a specified group of reactors.

The generation of this data for PWRs is conceptually straightforward but presents a significant data storage and handling problem. The computational work can be carried on at three levels of automation: completely unautomated, partially automated, completely automated. The choice of the level of automation depends on a balance of several considerations including the volume of computations to be performed, the cost of generating the computer programs, and the degree of reliability and flexibility sought in the system. Perhaps the best choice would be a combination of manual and machine computation. The straightforward lookup and interpolation operation in the table of isotopic composition vs exposure; and the simple bookkeeping operations could be done by hand. This would alleviate the requirement for large amounts of data storage. If the ^{236}Pu radiation contribution can be considered negligible compared to the contributions from the other plutonium isotopes, the data of Reference 1 can be used without any additional

calculations of isotope concentration. More tedious procedures like the isotopic spatial averaging could be done by auxiliary programs.

The generation of data for BWRs using a combination of FLARE and ISOCHECK would automatically produce reactor volume average isotopic concentrations for the BWR being analyzed. The saving simplification in the BWR analysis lies in the high degree of similarity between BWRs, especially the later off-the-shelf designs, as is shown in Table II. A large uncertainty in the characterization of BWRs is the control rod patterns used during the irradiation. If some nominal control configuration is chosen then perhaps the BWRs can be characterized by 3 or 4 FLARE - ISOCHECK calculations.

FIGURE 1. ZODIAC Calculated Pu239-Pu242 Isotopic Concentration vs Analytical Data

FIGURE 2. ZODIAC Calculated Pu236 Isotopic Concentration vs Analytical Data

FIGURE 3. ZODIAC Calculated U236, Np237, Pu238 Isotopic Concentration vs Analytical Data

FIGURE 4. FORT CALHOUN - Dissolved Boron Concentration Evaluation as a Function of Burnup

FIGURE 5. Plot of Burnup for FORT CALHOUN Versus U235

FIGURE 6. Plot of Pu236 Versus U235 for FORT CALHOUN

FIGURE 7. Plot of Pu238 Versus U235 for FORT CALHOUN

FIGURE 8. Plot of Pu239 Versus U235 for FORT CALHOUN

FIGURE 9. Plot of Pu240 Versus U235 for FORT CALHOUN

FIGURE 10. Plot of Pu241 Versus U235 for FORT CALHOUN

FIGURE 11. Plot of Pu242 Versus U235 for FORT CALHOUN

FIGURE 12. Plot of Np237 Versus U235 for FORT CALHOUN

FIGURE 13. Regional Power Versus Axial Position for a FORT CALHOUN Fuel Element

FIGURE 14. Regional Exposure Versus Axial Position for a FORT CALHOUN Fuel Element

FIGURE 15. FORT CALHOUN - PWR Average Exposure Versus Position Along Rod

FIGURE 16. FORT CALHOUN - PWR Pu236 Concentration Versus Position Along Rod at 28,000 MWD/T Average

FIGURE 17. FORT CALHOUN - PWR Pu238 Concentration Versus Position Along Rod at 28,000 MWD/T Average

FIGURE 18. FORT CALHOUN - PWR Pu239 Concentration Versus Position Along Rod at 28,000 MWD/T Average

FIGURE 19. FORT CALHOUN - PWR Pu240 Concentration Versus Position Along Rod at 28,000 MWD/T Average

FIGURE 20. FORT CALHOUN - PWR Pu241 Concentration Versus Position Along Rod at 28,000 MWD/T Average

FIGURE 21. FORT CALHOUN - PWR Pu242 Concentration Versus Position Along Rod at 28,000 MW./T Average

FIGURE 22. FORT CALHOUN - PWR Exposure Versus Position Along Rod

FIGURE 23. FORT CALHOUN - PWR Pu236 Concentration Versus Position Along Rod at 17055 MWD/T Average

FIGURE 24. FORT CALHOUN - PWR Pu238 Concentration Versus Position Along Rod at 17055 MWD/T Average

FIGURE 25. FORT CALHOUN - PWR Pu239 Concentration Versus Position Along Rod at 17055 MWD/T Average

FIGURE 26. FORT CALHOUN - PWR Pu240 Concentration Versus Position Along Rod at 17055 MWD/T Average

FIGURE 27. FORT CALHOUN - PWR Pu241 Concentration Versus Position Along Rod at 17055 MWD/T Average

FIGURE 28. FORT CALHOUN - PWR Pu242 Concentration Versus Position Along Rod at 17055 MW'D/T Average

ACKNOWLEDGEMENTS

The author is indebted to R. Holeman for his assistance especially in performing the ZODIAC calculations, to E. Reppond especially for her help on the reactor survey, to R. Matsen, J. Kutcher and N. Carter for many helpful suggestions on organization and content, to R. Sharp and R. McBride for technical assistance and to F. Totten and L. McKnight for typing the text.

REFERENCES

1. Theoretical Analysis of Plutonium Buildup and Uranium Depletion in Pressurized Water and Boiling Water Power Reactors. Final Report on Agency Research Contract 513/RB, Pacific Northwest Laboratories, Richland, Washington, August 1968.
2. R. H. Holeman and D. D. Matsumoto, ZODIAC (2+2) A Revision to ZODIAC 2, BNWL-459, Pacific Northwest Laboratory, Richland, Washington, May 1967.
3. R. J. Nodvik, Evaluation of Mass Spectrographic and Radiochemical Analysis of Yankee Core I Spent Fuel, WCAP-6068. Westinghouse Electric Corporation, Pittsburg, Pennsylvania, March 1966.
4. D. D. Matsumoto and G.J. Busselman, ZODIAC 2 User's Manual, HW-83577 General Electric Company, Richland, Washington, October 1964.
5. D. D. Matsumoto and R.H. Holeman, Reactor Theory and Code Development, BNWL-775, Pacific Northwest Laboratory, Richland, Washington, July 1968.
6. D. R. Skeen and W. L. Purcell, HTH-Computer Overlay of Programs HRG-THERMOS/BATTELLE-HFN. BNWL-634, Pacific Northwest Laboratory, Richland, Washington, February, 1968.
7. J. L. Carter, HRG3: A Code for Calculating the Slowing-Down Spectrum in the P₁ or B₁ Approximation, BNWL-1432, Pacific Northwest Laboratory, Richland, Washington, June, 1970.
8. C. L. Bennett and W. L. Purcell, BRT-I: Battelle-Revised-THERMOS-I. Pacific Northwest Laboratory, Richland, Washington, June, 1970.
9. J. R. Lilley, Computer Code HFN-Multigroup, Multiregion Neutron Diffusion Theory in One Space Dimension, HW-71545, General Electric Company, Richland, Washington, November, 1961.
10. G. J. Busselman, CLERK-A Cross Section Averaging Routine, HW-75521, General Electric Company, Richland, Washington, November, 1963.
11. B. H. Duane, Time-Variant Isotopic Transmutation GE-HL Program ALCHEMY, HW-80020, General Electric Company, Richland, Washington, December, 1963.
12. R. P. Matsen, An Analysis of Yankee-Rowe Burnup Data, BNWL-1122 Pacific Northwest Laboratory, Richland, Washington, July 1969.
13. R. P. Matsen, DBUFIT-1, A Least Squares Analysis Code for Nuclear Burnup Data, BNWL-1396, Pacific Northwest Laboratory, Richland, Washington, May 1970.

14. D. L. Delp, D. L. Fischer, J. M. Harriman, M. J. Stidwell, FLARE-A Three-Dimensional Boiling Water Reactor Simulator, GEAP-4598, July 16, 1964.
15. R. Kern, A. Forbes, T. Hencey, CETIC: A Computer Code Package Heavy Isotope Content in Reactors for use with Isochek Method, CEND-300, May 1967.

APPENDIX I

DESCRIPTION OF THE 1969 EVALUATION CROSS-SECTION DATA

APPENDIX IDESCRIPTION OF THE 1969 EVALUATION CROSS SECTION DATA

The 1969 cross-section evaluation was performed by B. Leonard. It contains corrections to existing cross-section data for many isotopes which are important to this study. In the case of ^{236}Pu and ^{242}Cm , data files were created for the first time on the BNW Master Library. Descriptions of the 1969 evaluation for 8 of the isotopes used in this study are as follows:

U-235

The recommended data file in the BNWML (Battelle Northwest Master Library), for ^{235}U is isotope number 414 labeled ^{235}W . This file contained errors in cross section values of ~1% for neutron energies below 0.01 eV. These errors apparently occurred when the files were renormalized to the 1965 IAEA recommended 2200m/s values.⁽¹⁾ The errors were corrected to give the cross section shapes which were originally intended. The thermal cross sections for ^{235}U have since been re-evaluated⁽²⁾ and should be updated again.

U-236

The thermal, resolved resonances, and unresolved resonance regions for ^{236}U were re-evaluated. The resonance description was based on recent precision capture measurements reported by Carlson.⁽³⁾ The 2200 m/s capture cross section was evaluated to be $5.28 \pm 0.25\text{b}$ based primarily on the value $5.1 \pm 0.25\text{ b}$ reported by Carlson and the value $5.5 \pm 0.3\text{b}$ deduced from an integral irradiation by Cabell, et al.⁽⁴⁾ The shape of the thermal cross

section was calculated from the positive-energy resonance parameters plus the negative-energy resonance parameters deduced by Carlson⁽³⁾ which were slightly modified to produce the chosen value of 2200 m/s cross section. The previous BNWML 2200 m/s value was 6.0 barns.

Np-237

A complete reevaluation of the ^{237}Np cross section to energies of 15 MeV was made. The previous BNWML version was inadequate and the original ENDF/B version⁽⁵⁾ contained several bad estimates. The results of the present evaluation of the most importance to the present function are summarized here.

No satisfactory existing evaluation of the thermal cross section could be found. The previous BNWML version prescribed a $1/v$ behavior of the capture cross section. The ENDF/B version⁽⁵⁾ increased faster than $1/v$ by about five percent. The ENDF/B evaluation stated that the only existing energy dependent total cross section data below 0.1 eV were due to a MTR crystal spectrometer measurement of Smith, et. al.⁽⁶⁾ Actually, a later set of measurements was made at the MTR by Cline, et al.⁽⁷⁾ and these measurements were considered by the Director of that program to supersede the earlier measurements of Smith, et. al.⁽⁸⁾ In addition, total cross section data in the thermal range were presented by a Russian group⁽⁹⁾ at the Geneva Conference. Strangely, neither the thermal data of Cline, et. al. or the Russian data have ever appeared in any of the issues of⁽¹⁰⁾ BNL-325 although both are cited in CINDA.⁽¹¹⁾ The data

of Cline, et. al. show a decrease from $1/v$ of some 30 percent in the thermal region. The Russian data show a somewhat larger decrease and a cross section value some 60 percent larger than the Cline data. Since the absolute values of the Cline data are more consistent with integral measurements these data were fitted to obtain the thermal cross-section shape in this evaluation. The fit required the additional of two negative-energy resonances and gives a 2200 m/s capture cross section of 169 barns. The uncertainty of the value and shape of the thermal cross section is about ± 10 percent which is the accuracy quoted⁽⁸⁾ for the data of Cline, et al.

All of the unmeasured partial cross sections for high energy neutrons given in the ENDF/B evaluation were based on an optical-model calculation by Goldman.⁽¹²⁾ However, the ENDF/B evaluators interpreted the total non-elastic cross section calculated by Goldman as the neutron inelastic scattering cross section. Thus an extensive reevaluation of the high-energy cross section was required. Of most importance to the present function was the evaluation of the $n, 2n$ cross section which leads to the highly radio-active contaminant ^{236}Pu . The presently evaluated $n, 2n$ cross section is based on the statistical calculation of Pearlstein⁽¹³⁾ which was used in the original ENDF/B evaluation.⁽⁵⁾ However, in the present evaluation Pearlstein's values have been adjusted for competition with other partial processes and rescaled to better agree with a single measured value at 14.5 MeV.⁽¹⁴⁾

Since the present evaluation, more extensive data in the resolved resonance region has become available and this region should be reevaluated.

Pu-236

^{236}Pu is a highly radioactive contaminant of long-burned nuclear fuel. The only neutron cross section information available is a measurement of the thermal fission cross section of 170 ± 35 barns.⁽¹⁵⁾ Expectation values of thermal alpha (~ 0.5) and average resonance parameters were estimated from nuclear systematics and used to generate a complete data file of capture and fission for this isotope. The average values of the capture cross section and the fission cross section beyond the thermal region are probably uncertain to about fifty percent.

Pu-238

The cross sections of ^{238}Pu were reevaluated in the thermal and resonance region. The same sources of data were used as were used in the ENDF/B evaluation.⁽¹⁶⁾ It appears, however, that the ENDF/B evaluation used a preliminary version of the results of the total cross section measurements of Young, et. al.⁽¹⁷⁾ Thus the resonance parameters of this evaluation differ somewhat from those of ENDF/B. There is a further arbitrariness about the derivation of the resonance parameters since Young et. al. derived their parameters on the implicit assumption that the fission widths were zero whereas they are a significant fraction of the capture width.⁽¹⁸⁾ A different treatment of the parameters of the negative energy resonances in the present evaluation produces better agreement than ENDF/B with the fission cross section measurements of Gerasimov⁽¹⁹⁾ for energies below the first positive energy resonance.

Pu-242

The original version of the ENDF/B gave a 2200 m/s capture cross section of 30 barns.⁽¹⁶⁾ The choice of this value was based on an erroneous best value of 30 ± 2 b given in BNL-325 (1958)⁽²⁰⁾ based on a preliminary value of Butler and Lounsbury⁽²¹⁾ given as 30.2 ± 1.1 barns. The value of Butler, et. al. had actually been changed⁽²²⁾ in 1957 to 18.6 ± 0.8 b based on a reevaluation of the resonance flux in that measurement. The previous version of the BNWML gave a 2200 m/s capture cross section of 26 barns based on preliminary total cross section data of Young.⁽²³⁾ Subsequent measurements by Young and Reeder⁽²⁴⁾, however, indicated that these data could not be meaningfully interpreted because of postulated experimental problems. In the present evaluation the 2200 m/s capture cross section was chosen to be 20 barns which is slightly larger than the value of Butler, et. al.⁽²²⁾ in recognition of the fact that all of the other integral measurements and differential measurements, although of lower accuracy, yield higher values. The accuracy of the thermal value is estimated to be about 10 percent.

The resonance parameter description of the present evaluation is essentially the same as given in ENDF/B⁽¹⁶⁾ except for the addition of a negative-energy level required to produce the 2200 m/s capture value of 20 barns.

Am-241

The shape of the thermal cross section was reevaluated. The shape given in the previous BNWML was obtained by adding a $1/v$ contribution to the positive-energy levels' contribution to obtain the desired

normalization. The shape given in ENDF/B⁽²⁵⁾ was based on the total cross-section measurements of Adamchuk, et al.⁽⁹⁾ which are the only measurements in the thermal region. The ENDF/B shape, however, was defined by only six points below 0.14 eV which resulted in an irregular shape and an unrealistic shape for energies below 0.01 eV. In the present evaluation, the low energy total cross-section data⁽⁹⁾ were reanalyzed to derive parameters for an additional level at negative energies. The shape of the thermal absorption cross section in the present file differs by some 25 percent from that given in the previous BNWML listing. The resonance parameter file is unchanged except for the addition of the negative-energy resonance.

The higher-energy files given in ENDF/B⁽²⁵⁾ are based on the same erroneous interpretation of the optical model calculations of Goldman⁽¹²⁾ as was made⁽⁵⁾ for ²³⁷Np. Neither the BNWML nor ENDF/B representations are valid estimates for fast reactor calculations.

Cm-242

No meaningful data files previously existed for ²⁴²Cm. The only known nuclear data consists of effective pile values of 20 ± 10 barns for capture⁽²⁶⁾ and < 5 barns for fission⁽²⁶⁾ and a recent measurement of 150 ± 40 b for the capture resonance integral.⁽²⁷⁾ These values and nuclear systematics were used to construct data files for the thermal, resolved resonance and unresolved resonance region for the BNWML. The uncertainties in effective cross section values calculated from these files are the order of 50 percent.

CROSS SECTION REFERENCES

1. C. H. Westcott, K. Ekberg, G. C. Hanna, N. J. Pattenden, S. Sanatani and P. M. Attree, *At. Energy Review*, 3, 3 (1965).
2. B. R. Leonard, Jr., Data provided for ENDF/B-Version 2 (1969).
3. A. D. Carlson, Unpublished data.
4. M. J. Cabell, T. A. Eastwood and P. J. Campion, *J. Nucl. Energy* 7, 81, (1958).
5. J. R. Smith, R. A. Grimesey, IN-1182, ENDF-116 (May 1969).
6. M. S. Smith, R. R. Smith, E. G. Joki and J. E. Evans, *Phys. Rev.* 107, 525 (1957).
7. J. E. Cline, E. H. Magleby, and W. H. Burgus, *Bull. Am. Phys. Soc.* 4, 207 (1959).
8. Letter from J. E. Evans, Director, Nuclear Physics Branch, Phillips Petroleum Co., Idaho Falls, Idaho, ref. Ev-103-59A, to M. R. Brussel, BNL, dated Aug. 26, 1969.
9. Y. B. Adamchuk, V. F. Gerasimov, B. V. Yefimos, V. S. Zenkevick, V. I. Mostovoi, M. I. Pezner, A. A. Chernyshov and A. P. Tsitovich, *Peaceful Uses of Atomic Energy*, Geneva, Vol. 4, pp 216-223 (1955).
10. J. R. Stehn, M. D. Goldberg, R. Wiener-Chasemen, S. F. Mughabghab, B. A. Magurno, and V. M. May, BNL-325, wnd Ed., Suppl. 2 (Feb. 1965).
11. CINDA (Computer Index of Neutron Data), USAEC Div. of Tech. Inf. Extension (May 1969).
12. D. T. Goldman, *Trans. Am. Nucl. Soc.* 7, 84 (1964).
13. S. Pearlstein, *Nucl. Sci. Engr.* 23, 238 (1965).
14. J. L. Perkins and R. F. Coleman, *J. Nucl. Energy* 14, 69 (1961).
15. J. E. Gindler, J. Gray, Jr. and J. R. Huizenga, *Phys. Rev.* 115, 1271 (1959).

16. C. L. Dunford and H. Alter, NAA-SR-12271 (May 1967) and NAA-SR-12271 Suppl. 1 (Sept. 1967).
17. T. E. Young, F. B. Simpson, J. R. Berreth and M. S. Coops, IN-1026 (Mar. 1967).
18. W. F. Stubbins, C. D. Bowman, G. F. Auchampaugh and M. S. Coops, Phys. Rev. 154, 1111 (1967).
19. V. F. Gerasimov, Sov. J. Nucl. Phys. 4, 706 (1967).
20. D. J. Hughes and R. B. Schwartz, BNL-325, 2nd Edition (July 1, 1958).
21. J. P. Butler and M. Lounsbury, AECL report PR-CM-3 (1955).
22. J. P. Butler, M. Lounsbury and J. S. Meritt, Can. J. Phys. 35, 147 (1957).
23. T. E. Young, IDO-17191, p. 25 (1965).
24. T. E. Young and S. D. Reeder, IN-1132 (June, 1968).
25. J. R. Smith and R. A. Grimesey, Idaho Nuclear Corp. unpublished "Comments on Evaluation of Np-237, Am-241 and Am-243," (Nov. 11, 1966).
26. D. J. Hughes and R. B. Schwartz, BNL-325, 2nd Ed. (1958).
27. R. P. Shumann, Unpublished data.

DISTRIBUTION

No. of
Copies

OFFSITE

1	<u>AEC Chicago Patent Group</u> G. H. Lee
8	<u>AEC Division of Reactor Development and Technology</u> Director RDT Asst. Dir. for Project Management Chief, Water Reactors Branch (2) Asst. Dir. for Reactor Technology Chief, Reactor Physics Branch (2) Chief, Fuels Recycle Branch
1	<u>AEC Division of Reactor Licensing</u> R. E. Baker
3	<u>AEC Division of Technical Information Extension</u>
4	<u>Argonne National Laboratory</u> R. Avery C. H. Bean R. E. Machery W. B. Seefeldt
1	<u>Babcock and Wilcox</u> H. M. Jones
1	<u>Bhabha Atomic Research Centre</u> Theoretical Physics Section/RED Central Complex Building Trombay, Bombay-85, India S. R. Dwivedi
1	<u>Brookhaven National Laboratory</u> J. Chernick
3	<u>Combustion Engineering</u> W. P. Chernock R. Harding S. Visner

No. of
Copies

1	<u>CNEN - Centro Studi-Nucleaire</u> Casaccia, Rome, Italy Ugo Farinelli
3	<u>Edison Electric Institute</u> 750 Third Avenue New York, New York 10017 John J. Kearney Harvey Wagner George Watkins
2	<u>E. I. du Pont de Nemours & Co., Inc., SRL</u> G. Dessauer H. Honeck
1	<u>ENEL</u> Via G. B. Martini (Piazza Verdi) Rome, Italy Mr. Paoletti Gualandi
3	<u>General Electric Co., San Jose</u> D. L. Fischer S. Levy R. B. Richards
4	<u>General Electric Co., Vallecitos Atomic Lab.</u> B. F. Judson T. M. Snyder R. E. Harris E. Kurtz
1	<u>Idaho Nuclear Corporation</u> R. Grimesey
1	<u>Japan Atomic Energy Institute</u> Tokaimura, Ibarakiken, Japan Shojiro Matsuura, JPDR-TCA
2	<u>Jersey Nuclear Enterprises</u> Bellevue, Washington R. L. Dickeman W. L. Hampson, Jr.
1	<u>Kerr-McGee</u> Dr. Frank Pittman Dr. I. D. Thomas

No. of
Copies

- 2 Nuclear Fuel Service
Wheaton, Maryland
J. D. Rollins
E. Wick
- 2 Nuclear Materials and Equipment Corp.
C. S. Caldwell
K. Puechl
- 1 NUKEM
D-645, HANAU
POSTFACH 869
Germany
Mr. Wolfgang K. L. Jager
- 1 Pakistan Institute of Nuclear Sci. & Tech.
P. O. Nilore
Rawalpindi, Pakistan
M. A. Mannan
- 1 Power Reactor & Nuclear Fuel Development Corp.
9-13, 1-chome, Akasaka
Minato-ku, Yokyo, Japan
Setsuo Kobayashi
- 1 S. C. K. - C. E. N.
MOL-DONK
Belgium
Dr. H. Vanden Broeck
BRL.
- 2 United Nuclear Corporation
T. B. Holden
J. R. Tomonto

No. of
Copies

6 Westinghouse Electric Corporation
 R. J. French
 R. S. Miller
 P. M. Murray
 J. Nardi
 R. E. Olsen
 J. B. Roll

ONSITE - HANFORD

1 AEC Chicago Patent Group
 R. K. Sharp

4 RDT Assistant Director for Pacific Northwest Programs
 W. E. Fry
 P. G. Holsted
 J. B. Kitchen
 T. A. Nemzek

2 AEC Richland Operations Office
 H. A. House
 C. L. Robinson

71 Battelle-Northwest

F. W. Albaugh	M. D. Freshley
C. L. Bennett	G. L. Gelhaus
L. W. Brackenbush	S. Goldsmith
S. H. Bush	R. L. Gulley
G. J. Busselman	H. J. Haverfield
J. L. Carter	H. L. Henry
N. E. Carter (2)	U. P. Jenquin
D. E. Christensen	D. A. Kottwitz
F. G. Dawson	J. W. Kutcher
D. R. de Halas	B. R. Leonard
D. E. Deonigi	R. C. Liikala
J. R. Devine	C. W. Lindenmeier
L. G. Faust	
L. J. Federico	
J. C. Fox	

No. of
Copies

R. P. Matsen
J. L. Maryott
D. F. Newman
D. R. Oden
D. L. Prezbindowski (20)
R. H. Purcell
W. D. Richmond
L. C. Schmid
R. I. Smith

V. O. Uotinen
H. H. VanTuyt
A. D. Vaughn
N. G. Wittenbrock
W. C. Wolkenhauer
M. G. Zimmerman
BNW Legal (2)
Technical Information (5)
Technical Publications (2)

25

WADCO Corporation

H. J. Anderson
J. M. Atwood
J. G. Bradley
J. J. Cadwell
J. A. Christensen
E. A. Evans
G. A. Halseth
E. N. Heck
R. E. Heineman

P. L. Hofmann
J. N. Judy
R. E. Lerch
R. B. Richman
R. C. Smith (5)
L. D. Turner
K. L. Young
WADCO Document Control (5)