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DRESDEN 1 PLUTONIUM RECYCLE PROGRAM FINAL REPORT

by
STUART D. BRESNICK

Prepared under
Contract EY-76-C-02-2210
for the Chicago Operations Office
Department of Energy

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DATE PUBLISHED: JANUARY 1980

GENERAL ATOMIC COMPANY

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1. INTRODUCTION

This is the final report on the Dresden 1 Plutonium Recycle Demonstration Program. It covers the work performed from July 1, 1978 to completion, which includes in-pool inspection of two fuel assemblies, removal of two fuel rods, and post-irradiation examination (PIE) of six fuel rods.

Appendix A describes the inspection and rod removal operations, and Appendix B describes the PIE work.

2. SUMMARY OF PROGRAM

This work was carried out under a program initiated by United Nuclear Corporation and Commonwealth Edison Company in 1967. Fuel design and fabrication was done by Gulf Nuclear Fuels Company (formerly Gulf United Nuclear Fuels Corporation and United Nuclear Corporation).

Eleven fuel assemblies, each containing nine mixed oxide ($\text{PuU}_2\text{-UO}_2$) fuel rods, were designed, fabricated, and delivered as part of the Batch 7 reload fuel for Dresden 1. Irradiation of this fuel began in January 1970, and the last mixed oxide assemblies were removed from the core in January 1977.

Design and manufacture of the fuel are described in Refs. 1 through 5.

Performance of the assemblies was satisfactory. Details of power density and exposure history are described in the progress reports listed in Refs. 6 through 13.

After cycle 7 ($\sim 11,000$ MWd/MT heavy metal average assembly exposure), all 11 assemblies were visually inspected (by underwater TV) and sipped for gas leakage. All assemblies were found to be in good condition and were returned to the core.

At the end of cycle 8 (October 1973), all 11 assemblies were sipped. Six were found to be leaking and were not loaded for the next cycle. Average assembly exposure at this time was $\sim 15,900$ to $17,500$ MWd/MT heavy metal. Visual inspection (underwater TV) of the assemblies showed several end-plug weld failures and cladding blisters. These defects were not unique to the mixed oxide fuel; the UO_2 fuel also exhibited such defects.

At the end of cycle 9 (September 1975), two assemblies were visually inspected, and no defects or abnormalities were found. Average assembly exposure was ~19,000 to 20,000 MWd/MT heavy metal.

At this time, four mixed oxide fuel rods were removed, one from each of two assemblies removed at the end of cycle 8 and the end of cycle 9.

At the end of cycle 10 (June 1977), the remaining mixed oxide assemblies were removed from the core due to high exposure (~24,000 to 25,000 MWd/MT heavy metal average assembly exposure). At this time, a visual inspection was made of two assemblies, and one mixed oxide rod was removed from each of these assemblies. The inspection showed no defects or abnormalities. Details of this inspection and rod removal are described in Appendix A.

The final phase of this program was the PIE of the six fuel rods, representing three stages of fuel burnup. The PIE work was done by Babcock and Wilcox, in accordance with the scope described in the Department of Energy contract. Appendix B contains the entire Babcock and Wilcox PIE report.

REFERENCES

1. "Dresden Plutonium Demonstration Program - Proof-Test Facility - Experiments and Results," DOE Report GU-5285, Gulf United Nuclear Fuels Corporation, October 1972.
2. "PLATR Critical Experiments on Mixed-Oxide Fuel Assemblies for BWR's and Comparison with Calculations," DOE Report GU-5289, Gulf United Nuclear Fuels Corporation, January 1973.
3. "Nuclear Design, Economics and Safety Analysis of the Dresden I Mixed Oxide Demonstration Assemblies," DOE Report GU-5291, Gulf United Nuclear Fuels Corporation, December 1972.
4. "Mechanical Design of Dresden I Mixed Oxide Demonstration Assemblies," DOE Report GU-5292, Gulf United Nuclear Fuels Corporation, March 1973.
5. "Mixed Oxide Fuel Pellet Fabrication and Fuel Rod Assembly for Dresden I, Batch 7 Reload," DOE Report GU-5308, Gulf United Nuclear Fuels Corporation, January 1974.
6. "Semiannual Report for Period Ending June 30, 1972," DOE Report GU-5287, Gulf United Nuclear Fuels Corporation, January 1973.
7. "Semiannual Report for Period Ending December 31, 1972," DOE Report GU-5290, Gulf United Nuclear Fuels Corporation, January 1973.
8. "Semiannual Report for Period Ending June 30, 1973," DOE Report GU-5304, Gulf United Nuclear Fuels Corporation, July 1973.
9. "Semiannual Report for Period Ending December 31, 1973," DOE Report GU-5309, Gulf United Nuclear Fuels Corporation, January 1974.
10. "Dresden 1 Plutonium Recycle Report for the Period January 1, 1974 through June 30, 1975," DOE Report GA-A13813, General Atomic Company, December 1975.
11. "Dresden 1 Plutonium Recycle Program Report for the Period July 1, 1975 through December 31, 1975," DOE Report GA-A14106, General Atomic Company, July 1976.

12. "Dresden 1 Plutonium Recycle Program Report for the Period January 1, 1976 through June 30, 1976," DOE Report GA-A14302, General Atomic Company, January 1977.
13. "Dresden 1 Plutonium Recycle Program Report for the Period July 1, 1976 through June 10, 1977," DOE Report GA-A14989, General Atomic Company, January 1978.

APPENDIX A FUEL INSPECTION

A.1. SUMMARY

This report, prepared by Nuclear Energy Services, Inc. (NES) for General Atomic Company (GA), presents the results of the Plutonium-Bearing Fuel Assembly Visual Inspection and Fuel Rod Removal Programs performed by NES, at the Commonwealth Edison Company Dresden 1 Nuclear Power Station. Visual inspection of the two fuel assemblies revealed no structural defects or abnormalities which would affect the normal operation of the fuel assembly in subsequent cycles. One fuel rod was removed from each fuel assembly and stored in a fuel rod storage basket in the Dresden 1 fuel storage pool.

A.2. INTRODUCTION

During the week of January 9, 1978, NES performed nuclear field services for GA at the Dresden 1 Nuclear Power Station. Two plutonium-bearing fuel assemblies were visually inspected with binoculars, and one plutonium-bearing removable fuel rod was removed from each fuel assembly and stored in a storage basket.

A.3. PROCEDURES

Fuel handling, visual inspection, and fuel rod removal procedures were performed in accordance with NES Document No. 80A0707. All visual observations were recorded on visual inspection data sheets, which are included at the end of this report. Removable fuel rod transfers to the fuel rod storage basket were documented on the fuel rod storage basket key, which is also included at the end of this report.

A.4. EQUIPMENT

A.4.1. Visual Inspection Equipment

The fuel assemblies were supported horizontally on the Commonwealth Edison Company saw table in the Dresden 1 fuel storage pool. The visual inspections were performed with Sears binoculars, 7 x 35, Extra Wide Angle. The fuel assembly was supported 14 ft from the inspector.

A.4.2. Fuel Rod Removal Equipment

The GA removable fuel rod handling tool and fuel rod storage basket were used for fuel rod removal and storage. The two Commonwealth Edison Company sipper-strippers were used to support the fuel assembly and the fuel rod storage basket during fuel rod removal operations. All fuel assembly transfers in the pool were performed by Commonwealth Edison Company personnel using the fuel handling machine.

A.5. RESULTS

A.5.1. Visual Inspection Results

Detailed visual examination of the two plutonium-bearing fuel assemblies, UN-098 and UN-101, revealed no structural defects or abnormalities which would affect the normal operation of the fuel assemblies in subsequent cycles. The assemblies were free of any visual bowing; all spacer grids were found to be sound and uncocked.* Equal spacing and straightness of all visually accessible fuel rods were observed.

---

*The cocked spacer observed on Fuel Assembly No. UN-098 probably occurred during insertion of the fuel assembly into the saw table. The fuel handlers experienced some difficulty during the insertion of this fuel assembly into the saw table. The cocked spacer, however, did not preclude the proper installation of the shroud on the fuel assembly when reassembled after the visual inspection.

In addition, there were no visible indications of clad defects due to pellet-clad interactions. Crud patterns observed were considered normal.

The raw visual inspection data sheets are included at the end of this report.

A.5.2. Fuel Rod Removal Results

Forces required for breaking the fuel rods away from the spacers and forces required for withdrawal were considered normal for the two fuel rods removed. The two rods were removed and stored in the fuel rod storage basket, which will be stored in the Dresden 1 storage pool fuel assembly racks until further notice by GA. The fuel assemblies from which the two fuel rods were removed, their position within the fuel assemblies, and their present positions in the fuel rod storage basket are listed in Table A-1. The fuel rod storage basket key is included at the end of this report.

TABLE A-1
DISPOSITION OF FUEL RODS REMOVED FROM
PLUTONIUM-BEARING FUEL ASSEMBLIES

<u>Fuel Assembly No.</u>	<u>Position in Fuel Assembly</u>	<u>Position in Storage Basket</u>
UN-098	R2	5
UN-101	B2	6

NUCLEAR ENERGY SERVICES, INC. NES DIVISION VISUAL INSPECTION DATA SHEET	Fuel Assembly No. <u>UN-098</u>	Date <u>1-11-78</u>
	Exposure <u> </u>	Time Start <u>1010</u>
	Video Tape No. <u>NA</u>	Time Finish <u>1020</u>
	Index No. <u>NA</u> To <u>NA</u>	Approved <u> </u>
	Operator <u> </u>	Date <u> </u>
Inspector <u>LOU ZEZZA</u>		

	Check List	Yes	No
1. All Nuts Crimped Remarks		☒	☐
2. Rem. Rod Locked Remarks		☒	☐
3. All Springs Assembled Remarks		☒	☐
4. Spacer Damage Remarks		☐	☒
5. Free End Rods Bottomed-Out Remarks		☒	☐
6. Bowed Rods Remarks		☐	☒
7. Miscellaneous Observations Remarks		☒	☐

**SHADED AREAS REPRESENT CRUD PATTERNS -
UNSHADED AREAS ARE CRUD FREE.**

NUCLEAR ENERGY SERVICES, INC.
NES DIVISION

VISUAL INSPECTION DATA SHEET

Fuel Assembly No. UN-098

Exposure _____

Video Tape No. NA

Index No. NA To NA

Operator _____

Inspector LOU ZEZZA

Date 1-11-78

Time Start 0945

Time Finish 0955

Approved _____

Date _____

Rod Location No.

E6 E5 E4 E3 E2 E1

Side B

Check List

	Yes	No
1. All Nuts Crimped Remarks _____	☒	☐
2. Rem. Rod Locked Remarks _____	☒	☐
3. All Springs Assembled Remarks _____	☒	☐
4. Spacer Damage Remarks _____	☐	☒
5. Free End Rods Bottomed-Out Remarks _____	☒	☐
6. Bowed Rods Remarks _____	☐	☒
7. Miscellaneous Observations Remarks _____	☒	☐
<u>a. E4 NUT RAISED ~ 1/10" ABOVE GRID PLATE</u>		
<u>b. CRUD PATTERNS ARE SIMILAR TO SIDE A</u>		

NUCLEAR ENERGY SERVICES, INC.
NES DIVISION

VISUAL INSPECTION DATA SHEET

Fuel Assembly No. UN-098
Exposure _____
Video Tape No. NA
Index No. NA To NA
Operator _____
Inspector LOU ZEPPE

Date 1-11-78
Time Start 0925
Time Finish 0940
Approved _____
Date _____

Check List

	Yes	No
1. All Nuts Crimped Remarks	☒	☐
2. Rem. Rod Locked Remarks	☒	☐
3. All Springs Assembled Remarks	☒	☐
4. Spacer Damage Remarks	☐	☒
<u>SPACER NO. 5 COCKED ABOUT 1/8" IN</u> <u>DIRECTION SHOWN - PROBABLY OCCURRED DURING</u> <u>INSERTION INTO SAW TABLE ?</u>		
5. Free End Rods Bottomed-Out Remarks	☒	☐
6. Bowed Rods Remarks	☐	☒
7. Miscellaneous Observations Remarks	☒	☐
<u>a. D5 NUT LIFTED ~ 1/8" ABOVE GRID PLATE</u> <u>b. CRUD PATTERNS ARE SIMILAR TO SIDE A</u>		

NUCLEAR ENERGY SERVICES, INC.
NES DIVISION

VISUAL INSPECTION DATA SHEET

Fuel Assembly No. UN-098

Exposure -

Video Tape No. NA

Index No. NA To NA

Operator -

Inspector LOU ZEZZA

Date 1-11-78

Time Start 0900

Time Finish 0915

Approved -

Date -

Check List

	Yes	No
1. All Nuts Crimped Remarks	☒	☐
2. Rem. Rod Locked Remarks	☒	☐
3. All Springs Assembled Remarks	☒	☐
4. Spacer Damage Remarks	☐	☒
5. Free End Rods Bottomed-Out Remarks	☒	☐
6. Bowed Rods Remarks	☐	☒
7. Miscellaneous Observations Remarks	☒	☐
a. B3 NUT RAISED ~ 1/8" ABOVE GRID PLATE		
b. CRUD PATTERNS ARE SIMILAR TO SIDE A		

NUCLEAR ENERGY SERVICES, INC. NES DIVISION VISUAL INSPECTION DATA SHEET	Fuel Assembly No. <u>UN-101</u>	Date <u>1-11-78</u>
	Exposure <u>-</u>	Time Start <u>1115</u>
	Video Tape No. <u>NA</u>	Time Finish <u>1120</u>
	Index No. <u>NA</u> To <u>NA</u>	Approved <u>-</u>
	Operator <u>-</u>	Date <u>-</u>
	Inspector <u>LOU ZEZZA</u>	

Check List		Yes	No
1. All Nuts Crimped	Remarks	☒	☐
2. Rem. Rod Locked	Remarks	☒	☐
3. All Springs Assembled	Remarks	☒	☐
4. Spacer Damage	Remarks	☐	☒
5. Free End Rods Bottomed-Out	Remarks	☒	☐
6. Bowed Rods	Remarks	☐	☒
7. Miscellaneous Observations	Remarks	☒	☐
CRUD PATTERNS ARE SIMILAR TO SIDE A ON UN-098			

NUCLEAR ENERGY SERVICES, INC.
NES DIVISION

VISUAL INSPECTION DATA SHEET

Fuel Assembly No. UN-101

Exposure —

Video Tape No. NA

Index No. NA To NA

Operator —

Inspector LDU ZEZZA

Date 1-11-78

Time Start 1130

Time Finish 1135

Approved —

Date —

Rod Location No.

E6 E5 E4 E3 E2 E1

Side B

Check List

	Yes	No
1. All Nuts Crimped Remarks	☒	☐
2. Rem. Rod Locked Remarks	☒	☐
3. All Springs Assembled Remarks	☒	☐
4. Spacer Damage Remarks	☐	☒
5. Free End Hods Bottomed-Out Remarks	☒	☐
6. Bowed Rods Remarks	☐	☒
7. Miscellaneous Observations Remarks	☒	☐
CRUD PATTERNS ARE SIMILAR TO SIDE A		

 NUCLEAR ENERGY SERVICES, INC. NES DIVISION VISUAL INSPECTION DATA SHEET	Fuel Assembly No. <u>UN-101</u>	Date <u>1-11-78</u>
	Exposure <u> </u>	Time Start <u>1125</u>
	Video Tape No. <u>NA</u>	Time Finish <u>1130</u>
	Index No. <u>NA</u> To <u>NA</u>	Approved <u> </u>
	Operator <u> </u>	Date <u> </u>
	Inspector <u>LOU ZEZZA</u>	

	Check List	Yes	No
1. All Nuts Crimped Remarks		☒	☐
2. Rem. Rod Locked Remarks		☒	☐
3. All Springs Assembled Remarks		☒	☐
4. Spacer Damage Remarks		☐	☒
5. Free End Rods Bottomed-Out Remarks		☒	☐
6. Bowed Rods Remarks		☐	☒
7. Miscellaneous Observations Remarks		☒	☐
<u>CRUD PATTERNS ARE SIMILAR TO SIDE A</u>			

NUCLEAR ENERGY SERVICES, INC.
NES DIVISION

VISUAL INSPECTION DATA SHEET

Fuel Assembly No. UN-101
Exposure
Video Tape No. NA
Index No. NA To NA
Operator
Inspector LOU ZEZZA

Date 1-11-78
Time Start 1120
Time Finish 1125
Approved
Date

Check List

	Yes	No
1. All Nuts Crimped Remarks	☒	☐
2. Rem. Rod Locked Remarks	☒	☐
3. All Springs Assembled Remarks	☒	☐
4. Spacer Damage Remarks	☐	☒
5. Free End Rods Bottomed-Out Remarks	☒	☐
<u>A6 RAISED ~ 1/16"</u>		
6. Bowed Rods Remarks	☐	☒
7. Miscellaneous Observations Remarks	☒	☐
<u>2. B3 & B4 NUTS RAISED ~ 1/16"</u>		
<u>b. CRUD PATTERNS ARE SIMILAR TO SIDE A</u>		

NUCLEAR ENERGY SERVICES, INC.

ORIGINAL SIGNED BY
LOUIS J. ZEZZA 1-11-78

ORIENTATION
FEATURE

FUEL ROD STORAGE BASKET KEY

APPENDIX B
POST-IRRADIATION EXAMINATION

Babcock & Wilcox

POST-IRRADIATION EXAMINATION OF MIXED OXIDE FUEL RODS

**RESEARCH AND DEVELOPMENT DIVISION
LYNCHBURG RESEARCH CENTER
Lynchburg, Virginia**

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November 1979

POST-IRRADIATION EXAMINATION OF
MIXED OXIDE FUEL RODS

by

Vishnu Subrahmanyam,
Todd L. Hardt,
Joyce St. John

LRC

Prepared for

General Atomic Company
Under G.A.C. P.O. #619004
B&W CRD 3038

Submitted by

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ACKNOWLEDGMENT

The assistance of Harold Collins, Dan Miller, and the Hot Cell Operations crew in conducting the gamma scans and rod sectioning is gratefully acknowledged.

SUMMARY

Post-irradiation examination of three pairs of mixed oxide fuel rods was conducted by nondestructive gamma scan and radiochemical analysis methods.

The gross gamma scans indicated that there were no major changes in the physical form of the fuel pellets due to irradiation. Axial migration of fission products was ruled out based on the absence of areas of high activity, particularly at the interpellet regions.

It is known that relative fuel burnup profiles are represented by the concentrations of primary fission products. Relative fuel burnup profiles were constructed with the aid of axial distributions of fission products determined by analyzing gamma spectra recorded at different axial locations.

The distribution of long-lived ^{137}Cs ($T_{1/2} = 30.1$ y) representing the fuel burnup profile for the entire irradiation period indicated the maximum burnup to be located between 100 and 135 cm from the bottom of the fuel stack.

Axial distributions of ^{106}Ru ($T_{1/2} = 368$ d) and ^{95}Zr ($T_{1/2} = 64.0$ d) represent fuel burnup (power rating) profiles during portions of irradiation and late-in-life, respectively. These profiles suggested higher rates of fuel burnup in regions away from the center of the fuel rods. The axial relative neutron flux profile deduced from the distribution of ^{134}Cs ($T_{1/2} = 2.06$ y) activity was different for each fuel rod.

Expressions were derived for time-averaged neutron fluxes and their squared values in terms of relative activity ratios of $^{134}\text{Cs}/^{137}\text{Cs}$ and the relative activity of ^{134}Cs , respectively.

The relative profiles of fuel burnup were translated into absolute profiles by utilizing radiochemical data from two positions on each rod.

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1. INTRODUCTION

Post-irradiation examination (PIE) of mixed oxide fuel elements is a requirement for determining the inventory of remaining fissionable material. Far more important reasons for a PIE are to develop an understanding of the characteristics of fuel burnup, and the identification and evaluation of all the parameters and their influences on controlled fission. Information of this nature is of great importance in the development and incorporation of scientific and technological improvements for fuel performance. For example, any major physical changes in fuel pellets and rods brought about by prolonged exposure to high neutron fields need thorough investigation prior to undertaking any corrective measures.

The above objectives can be achieved by both nondestructive and destructive analyses. The former is a passive method that is widely practiced¹⁻³ because of its desirable aspects. The destructive method, on the other hand, is based on proven chemical techniques but fraught with hazards because of the high radiation fields involved. Both of these techniques are employed in the present study to investigate irradiated mixed oxide fuel rods.

Six Dresden mixed oxide fuel rods were selected by the General Atomic Corporation for post-irradiation examination. Examination of these rods was performed by The Babcock & Wilcox Company's Lynchburg Research Center under Purchase Order 619004.

The six fuel elements were obtained in three pairs with different irradiation histories, each pair coming from irradiations ending in October 1973, August 1975, and December 1977.

The following analyses were requested by General Atomic Corporation for each of the six rods:

1. Gross gamma scan for calculation of axial burnup profile, a gamma scan for ^{95}Zr - ^{95}Nb to obtain a measure of late-in-life burnup and a gamma scan for ^{137}Cs . Analyze scans to determine burnup profile as a function of irradiation history and possible fission product migration.

2. Radiochemical analysis of two sections of each rod to determine:

- a. U and Pu isotopic abundances.
- b. ^{237}Np , ^{243}Am , and ^{241}Am content.
- c. Burnup in each section from the atomic ratios of ^{239}Pu to ^{238}U and ^{148}Nd to ^{238}U and absolute axial burnup profiles by correlating the gross gamma scan with these data.

This report presents the problem definition, analytical approach, and experimental details used to carry out the desired workscope. The results are then presented from both nondestructive and destructive methods and discussed on a comparative basis. Conclusions and recommendations for future work constitute the final section.

2. PROBLEM STATEMENT AND METHODS OF ANALYSIS

The neutron field set up by the self-sustained fission that is the source of energy for power generation in a nuclear reactor is also accompanied by some parasitic effects. These effects include neutron absorption by unused fuel and by stable fission products. Neglecting parasitic nuclear reactions, the fuel burnup rate depends on the neutron flux at any given time and location in a reactor and energy release is associated essentially with fission processes. Analysis of different portions of irradiated fuel rods for fission products and products from non-fission modes of fuel consumption provides information that is of vital importance in understanding fuel burnup and its proper utilization.

2.1. Analytical Approach

The mathematical treatment of nuclear fuel consumption is essentially identical to the study of a radioactive decay series starting from an initially pure parent provided an appropriate decay constant is assigned to the disappearance of fissionable atoms.⁴ In the decay series

where A represents a nucleus undergoing fission.

The decay constant λ_a is given by

$$\lambda_a = \Sigma(\phi_f)_i. \quad (2)$$

Σ is the microscopic fission cross-section (in cm^2) and $(\phi_f)_i$ is the neutron flux (neutrons/ $\text{cm}^2\text{-s}$) experienced by atoms of A at location i. In the case of mixed oxide fuel, Σ can be expressed following Tsuruta, et al.,⁵ as

$$\Sigma = n_1\sigma_1 + n_2\sigma_2 \quad (3)$$

where n_1 , n_2 , and σ_1 , σ_2 represent the mole fractions and fission cross sections, respectively, of the constituent fissionable fuel materials.

The quantities λ_b and λ_c in equation 1 are the radioactive decay constants of the daughter and granddaughter of fission. These are approximately modified, if necessary, for modes of disappearance of atoms of B and C in addition to radioactive decay. Thus,

$$\lambda_b = \lambda_b(\text{radioactive decay}) + \sigma_a \phi_i \quad (4)$$

where σ_a is the neutron absorption cross section of B and ϕ is the neutron flux at location i.

In the cases of fission products ^{137}Cs , ^{106}Ru , and ^{95}Zr , the neutron absorption term of equation 5 is negligible because of very small σ_a values and/or due to large radioactive decay constants. On the other hand, for a stable fission product such as ^{133}Cs , λ_b is totally due to neutron absorption.

The activities of B and C ($N_b \lambda_b$, $N_c \lambda_c$) following an irradiation for time, T, and at the end of a decay period, t, can be written⁴ in the following manner:

$$N_b \lambda_b = \frac{A_0 \lambda_a \lambda_b}{(\lambda_b - \lambda_a)} \left(e^{-\lambda_a T} - e^{-\lambda_b T} \right) e^{-\lambda_b t} \quad (5)$$

$$N_c \lambda_c = \frac{A_0 \lambda_a \cdot \lambda_b \cdot \lambda_c \cdot e^{-\lambda_c t}}{(\lambda_a - \lambda_b)(\lambda_b - \lambda_c)(\lambda_c - \lambda_a)} \left[(\lambda_c - \lambda_b) e^{-\lambda_a T} + (\lambda_a - \lambda_c) e^{-\lambda_b T} + (\lambda_b - \lambda_a) e^{-\lambda_c T} \right] \quad (6)$$

In equations 5 and 6, A_0 is the number of atoms of A at time $T = 0$ with $B = C = 0$.

From equation 5, the ratio of activities of B at locations i and j is given by

$$R_b = \frac{(N_b \lambda_b)_i}{(N_b \lambda_b)_j} = \frac{(A_0 \lambda_a)_i}{(A_0 \lambda_a)_j} \quad (7)$$

Thus, R_{137} , R_{106} , and R_{95} represent the relative fuel burnup values and relative activities of fission products ^{137}Cs , ^{106}Ru , and ^{95}Zr , respectively.

Again, from equation 6

$$R_c = \frac{(N_c \lambda_c)_i}{(N_c \lambda_c)_j} = \frac{(A_o \lambda_a \lambda_b)_i}{(A_o \lambda_a \lambda_b)_j} \quad (8)$$

In the specific case of ^{134}Cs , substituting for λ_a in equation 8,

$$R_{134} = \frac{(\phi_f)_i \phi_i}{(\phi_f)_j \phi_j} \approx \frac{\phi_i^2}{\phi_j^2} \quad (9)$$

The approximation in equation 9 implies that the fissioning nucleus and the fission product ^{134}Cs experience the same neutron flux at both locations i and j.

It can be seen then from equations 7 and 8 that

$$R_{134} = R_{137} \cdot \frac{\phi_i}{\phi_j} \quad (10)$$

The axial position corresponding to highest measured activity of each isotope was chosen as the reference position, location j, in the above expressions for R.

2.2. Experimental Approach

The following nondestructive and destructive methods of PIE were designed relative to the goals of this investigation:

2.2.1. Gamma Scan Method

This nondestructive approach to PIE is a passive method and was carried out in two stages. In the first stage, the activity of gamma rays above a certain minimum energy (~50 keV) and the gamma activity of one fission product (^{137}Cs) were monitored as the fuel rod axially traversed the scanning device. This "gross" scan was intended to detect any regions of high activity due to non-uniform fuel burnup or axial migration of fission products.

In the second part of this method, high resolution gamma spectra were recorded when the fuel rod was located so as to expose a well-defined segment of an axial position to the detector. The relative amounts of different fission products were determined by the area of characteristic full energy gamma ray photopeaks. These fission products remaining at their formation site represented the extent of fission at that location and hence portrayed relative

fuel burnup and power profiles. The relative concentration profiles of ^{137}Cs , ^{106}Ru , or ^{95}Zr and the relative activity ratio, R_{134}/R_{137} , were utilized in establishing time averaged neutron flux profiles at various axial positions.

The relative profiles of fuel burnup and power determined by the gamma scan method could be translated into time profiles by a measurement of absolute values along the fuel rod axis. For this purpose, transverse sections at two locations were obtained for each fuel rod and were analyzed radiochemically.

2.2.2. Destructive Method

Axial sections of fuel rods were dissolved and chemically analyzed. The radiochemical fractions were further analyzed by a mass spectrometer, and the following data were obtained:

1. U and Pu isotopic abundances.
2. ^{237}Np , ^{241}Am , and ^{243}Am amounts.
3. Atomic ratio $^{148}\text{Nd}/^{238}\text{U}$.

Isotopic abundances of U and Pu in the irradiated fuel can be utilized in the calculation of their respective depletions when the atomic composition of the fuel prior to irradiation is known.

The identifiable formation modes of ^{241}Am and ^{243}Am are the (n,γ) reactions. ^{237}Np , on the other hand, is formed predominantly by β decay of ^{237}U formed by successive capture of two neutrons by ^{235}U or the nuclear reaction $^{238}\text{U}(n,2n)^{237}\text{U}$. The modes of formation of these nuclides depend on the local neutron field. Their amounts, therefore, provide a means of evaluating neutron fluences.

The observed quantity of the shielded nuclide ^{148}Nd is a measure of total fissions at any location in the fuel, and the ratio $^{148}\text{Nd}/^{238}\text{U}$ can be used to determine the extent of fission.

The results of the destructive method can be compared with similar data from the gamma scan method. Since the relative values of fuel burnup and neutron flux were known for positions of the rods that were destructively analyzed, average absolute values for the unit on the relative scale can be calculated, and the relative neutron flux values translated into absolute values.

3. EXPERIMENTAL DETAILS

3.1. Gamma Scan Method

The experimental setup used consisted of positioning a fuel rod and viewing it through a double slit 1-m (~40-in.) collimator with a stationary Ge(Li) detector positioned outside the hot cell. As the rod moved vertically, the extent of its travel was indicated by a calibrated odometer and the gamma detector was exposed to a small segment of the rod.

The collimator slits were set at 1.0 mm (primary) and ~1.5 mm (scraper), respectively. A Ge(Li) detector with ~2 keV resolution at 1332 keV and 7% detection efficiency was employed for gamma detection and the detector signals were processed by ORTEC modular electronics.

Continuous gamma scans of gross gamma and ^{137}Cs activities were recorded with a dual-pen strip chart recorder. ^{95}Zr gamma activity was also recorded in one pair of rods in which the isotope was detected.

Gamma spectra were recorded for 1000 seconds (live time) at 10 equally spaced axial positions of each rod. These spectra were stored, analyzed, and the nuclides identified with a Nuclear Data 6620 system. The spectral analysis by the system was based on a proprietary computer code that determined the areas of full energy photopeaks to identify specific isotopes.

3.2. Rod Sectioning Operations

General Atomic selected the axial locations of the burnup samples based on previous operating history. Samples 2.5-cm (1-in.) long were taken at 75 and 80 cm (30 to 32 in.) and 243 and 248 cm (97 to 99 in.) from the bottom of the fuel rods for radiochemical analyses. Prior to sectioning, a detailed sample plan and specimen identification system were prepared for the rods. The rod sectioning diagrams are shown in Figures 1 and 2. The locations of the samples and waste sections were measured using a Lufkin rule graduated in 1/16 inch. Each rod was sectioned using a modified tubing cutter. Individual samples and waste sections were stored in marked containers. The 0.25-cm

(0.1-in.) thick wafers needed for analysis were subsequently cut from the 1 in. segments. Exact locations for each sample are shown in Table 1. To insure that a complete sample cross section was obtained for burnup analysis, the samples were impregnated with a mixture of acetone and Duco cement. Each sample was placed on a Buehler Isomet saw, measured and cut. A 0.25-cm (0.1-in.) thick wafer from each sample was placed in a marked vial to maintain identification.

3.3. Destructive Method

Each sample of fuel submitted for destructive burnup analysis consisted of a fuel rod slice including fuel approximately 3-5 mm thick. This sample was dissolved in a dilute aqua regia solution made from a 12:3:10 ratio of HNO_3 , HCl , and water, respectively. A portion of the dissolved mixture was diluted with 0.1N HCl and an aliquot of this final solution was used for analysis.

A spike containing known amounts of ^{233}U , ^{244}Pu , and ^{150}Nd was added to the aliquot of dissolved fuel. An unspiked fuel sample was also run in parallel to the spiked portion. The separation of U, Pu, and Nd used two sequential ion exchange columns operated at room temperature. The sample was fumed with HF and HClO_4 to convert the U and Pu to the VI oxidation state. The sample was transferred to the first AGMP-1 anion exchange column in 12N HCl . The effluent from this column containing Nd, other fission products, and Am were converted to the nitrate form. The Pu was eluted from the first column with an HI-HCl mixture and U was removed with 0.1N HCl . The fission products and Nd were loaded onto a second anion exchange column and Nd was isolated by chromatographic elution with a mixture of $\text{HNO}_3\text{-CH}_3\text{OH}$.

The U, Pu, and Nd fractions were then analyzed by mass spectroscopy to determine the isotopic content of each fraction and the chemical yield provided by the spike. These data were used to calculate the nuclear burnup.

Table 1. Identification and Location of
Samples for Burnup Analysis

<u>Rod No.</u>	<u>Sample</u>	<u>Axial location^(a)</u>	
		<u>cm</u>	<u>Inches</u>
UN 097	A	250.19	98.5
UN 097	B	77.47	30.5
UN 098	A	250.19	98.5
UN 098	B	77.47	30.5
UN 099	A	247.65	97.5
UN 099	B	80.01	31.5
UN 100	A	247.65	97.5
UN 100	B	80.01	31.5
UN 101	A	247.65	97.5
UN 101	B	80.01	31.5
UN 102	A	247.65	97.5
UN 102	B	80.01	31.5

(a) From bottom of fuel rod.

Figure 1. Rod Sectioning Diagram for Dresden Rods UN 097 and UN 098

Figure 2. Rod Sectioning Diagram for Dresden Rods UN 099,
UN 100, UN 101, and UN 102

4. RESULTS

4.1. Gamma Scan Method

Gross gamma scans of radioactive isotopes with gamma energies up to about 2.0 MeV and the single channel selected gamma activity of ^{137}Cs for the six rods are shown in Figures 3 and 4. Figure 5 contains the single-channel selected activity of $^{95}\text{Zr}/^{95}\text{Nb}$ for the pair of rods (UN 098 and UN 101) in which this activity was detected.

Tables 2 through 7 present the relative positional activities (R values) of characteristic gamma rays of isotopes ^{137}Cs , ^{134}Cs , and ^{106}Ru , along with R values for ^{95}Zr for rods in which this activity was detected. The axial positions where transverse sections were taken for radiochemical analysis are indicated by pointers. These R values, also shown in Figures 6 through 11, were computed by dividing the activity at each location by the highest observed activity. Since the decay corrections and attenuation factors for their growth during irradiation are identical for the same isotope regardless of its location in the fuel rod, the R values represent the relative numbers of atoms of the isotope at various axial positions.

The irradiation and decay history of the fuel rods relative to the time of counting is shown schematically in Figure 12. Periods corresponding to six half-lives of the shorter lived isotopes ^{106}Ru and ^{95}Zr are also shown in Figure 12. The period of growth of the detected activity during irradiation was determined (Figure 12), assuming the activity of these isotopes could be detected only for six half-lives.

4.2. Radiochemical Analyses

The isotopic analysis of ion exchange separated uranium, plutonium, and neodymium fractions of fuel rod samples are given in Table 8. The analyzed samples are also identified in Table 1. The number of atoms of each isotope is expressed relative to 10^6 atoms of ^{238}U present in the fuel after irradiation.

As explained earlier (see section 3.3), the amount of respective spike elements and their isotopic compositions were utilized in the calculations.

The number of atoms of U, Pu, and Nd isotopes given in Table 8 is based on a mass spectroscopic analysis of the respective fractions from ion exchange separations. The number of ^{241}Am and ^{237}Np atoms was evaluated by gamma counting aliquots of dissolved fuel samples. The intensities of 59.6 and 86.5 keV gamma rays in gamma spectra recorded with a calibrated gamma detection system were used in the calculations. The gamma ray characteristic of ^{243}Am (74.5 keV) was not identified in any of the gamma spectra recorded and analyzed. The minimum detectable intensity of this gamma ray with the detection system employed was 0.05 cps and corresponds to about one atom of ^{243}Am for every 10^6 atoms of ^{238}U in the dissolved sample for which the relative number of atoms of other isotopes are represented in Table 8.

The burnup was calculated from changes in the uranium and plutonium content as well as the fission product content (Nd) as described in ASTM methods E-244, E-267, and E-321.

The destructive analysis of a fuel sample yielded burnup as fissions per initial atom of uranium and plutonium. This value can be converted to gigawatt days per metric ton by multiplying by 960. The uncertainty in this conversion factor is 3%. The uncertainty in the measured values is approximately 3.5%. The overall error associated with the burnup expressed as GWD/mt is about 4.6%.

The initial isotopic abundances used in the calculations were the specification values for these assemblies, i.e., natural uranium, 71.7% ^{239}Pu , 20.6% ^{240}Pu , 6.4% ^{241}Pu , 1.0% ^{242}Pu , and 0.3% ^{238}Pu . The capture-to-fission ratios for the fissile isotopes were the default values in the General Electric burnup computer code.

Table 2. Relative Axial Concentrations of Fission Products in Fuel Rod UN 097

Axial position(a)	Isotope, E_{γ} (keV)	^{137}Cs 661.5	^{134}Cs 795.8	^{106}Ru 621.8	^{95}Zr 756.0			
cm	Inches	R_{137}	R_{134}	R_{106}	R_{95}	R_{134}/R_{137}	R_{106}/R_{137}	R_{95}/R_{137}
266.57	104.95	0.410	0.154	0.418		0.375	1.019	
250.57	98.65	0.590	0.362	0.591		0.614	1.003	
211.46	83.25	0.863	0.813	0.894		0.942	1.036	
182.75	71.95	0.928	0.909	1.000		0.979	1.077	
158.12	62.25	0.959	0.979	0.953		1.022	0.994	
131.70	51.85	0.974	0.980	0.998		1.007	1.025	
105.28	41.45	0.994	1.000	0.996		1.006	1.002	
66.42	26.15	1.000	0.966	0.982		0.966	0.982	
19.94	7.85	0.706	0.463	0.672		0.655	0.951	
0.38	0.15	0.399	0.117	0.362		0.293	0.908	
Maximum peak area		229,183	32,013	6,552				

(a) From bottom of fuel stack.

Table 3. Relative Axial Concentrations of Fission Products in Fuel Rod UN 102

Axial position (a)	Isotope, E_{γ} (keV)	^{137}Cs <u>661.6</u>	^{134}Cs <u>795.8</u>	^{106}Ru <u>621.8</u>	^{95}Zr <u>756.0</u>			
<u>cm</u>	<u>Inches</u>	<u>R_{137}</u>	<u>R_{134}</u>	<u>R_{106}</u>	<u>R_{95}</u>	<u>R_{134}/R_{137}</u>	<u>R_{106}/R_{137}</u>	<u>R_{95}/R_{137}</u>
267.46	105.30	0.456	0.183	0.437		0.401	0.959	
252.98	99.60	0.622	0.400	0.578		0.644	0.930	
213.36	84.00	0.924	0.894	0.838		0.968	0.907	
183.64	72.3	0.966	0.968	0.881		1.003	0.912	
157.99	62.2	0.991	0.980	0.904		0.990	0.912	
131.83	51.9	0.998	1.000	1.000		1.002	1.002	
106.68	42.00	1.000	0.951	0.891		0.951	0.891	
66.80	26.3	0.971	0.875	0.889		0.900	0.916	
19.81	7.80	0.671	0.408	0.682		0.609	1.016	
0.51	0.20	0.382	0.105	0.369		0.274	0.964	
Maximum peak area		237,462	35,486	7,338				

(a) From bottom of fuel stack.

Table 4. Relative Axial Concentrations of Fission Products in Fuel Rod UN 098

Axial position (a)	Isotope, E_{γ} (keV)	^{137}Cs 661.6	^{134}Cs 795.8	^{106}Ru 621.8	^{95}Zr 756.0			
cm	Inches	R_{137}	R_{134}	R_{106}	R_{95}	R_{134}/R_{137}	R_{106}/R_{137}	R_{95}/R_{137}
267.97	105.50	0.478	0.192	0.433	0.431	0.402	0.906	0.902
253.75	99.90	0.631	0.399	0.649	0.632	0.632	1.028	1.001
213.36	84.00	0.909	0.854	0.940	0.846	0.940	1.035	0.931
183.90	72.40	0.963	0.950	0.975	0.859	0.986	1.012	0.891
159.00	62.60	0.982	0.984	0.983	0.855	1.002	1.001	0.871
132.59	52.20	1.000	1.000	1.000	0.909	1.000	1.000	0.909
106.43	41.90	0.994	0.961	0.977	0.969	0.966	0.983	0.975
67.56	26.60	0.964	0.882	0.920	1.000	0.915	0.955	1.038
20.83	8.20	0.715	0.446	0.691	0.851	0.623	0.965	1.190
0.25	0.10	0.412	0.115	0.387	0.462	0.279	0.939	1.120
Maximum peak area		350,059	222,076	75,574	15,074			

(a) From bottom of fuel stack.

Table 5. Relative Axial Concentrations of Fission Products in Fuel Rod UN 101

Axial position (a)	Isotope, E_{γ} (keV)	^{137}Cs	^{134}Cs	^{106}Ru	^{95}Zr	R_{134}/R_{137}	R_{106}/R_{137}	R_{95}/R_{137}
		661.6	795.8	621.8	756.0			
cm	Inches	R_{137}	R_{134}	R_{106}	R_{95}			
268.43	105.68	0.452	0.182	0.461	0.498	0.402	1.020	1.102
252.86	99.55	0.623	0.410	0.661	0.715	0.653	1.053	1.139
212.98	83.85	0.910	0.861	0.925	0.926	0.947	1.016	1.018
182.75	71.95	0.974	0.969	0.978	0.950	0.994	1.003	0.975
159.13	62.65	0.997	1.000	1.000	0.955	1.003	1.003	0.958
132.21	52.05	0.996	0.986	0.894	0.975	0.990	0.898	0.979
106.55	41.95	1.000	0.939	0.959	0.917	0.979	0.959	0.917
67.18	26.45	0.923	0.944	0.883	1.000	0.951	0.889	1.007
21.21	8.35	0.743	0.504	0.683	0.788	0.678	0.919	1.061
0.13	0.05	0.403	0.121	0.402	0.450	0.301	0.998	1.117
Maximum peak area		339,878	177,540	66,069	11,661			

(a) From bottom of fuel stack.

Table 6. Relative Axial Concentrations of Fission Products in Fuel Rod UN 099

Axial position (a)	Isotope, E_γ (keV)	^{137}Cs	^{134}Cs	^{106}Ru	^{95}Zr			
		661.6	795.8	621.8	756.0			
cm	Inches	R_{137}	R_{134}	R_{106}	R_{95}	R_{134}/R_{137}	R_{106}/R_{137}	R_{95}/R_{137}
266.70	105.00	0.428	0.167	0.401		0.389	0.938	
252.22	99.30	0.595	0.361	0.558		0.607	0.938	
212.34	83.60	0.886	0.806	0.893		0.910	1.008	
181.86	71.60	0.950	0.925	0.982		0.973	1.033	
156.97	61.80	0.980	0.985	1.000		1.005	1.020	
130.30	51.30	0.996	1.000	0.945		1.004	0.978	
104.65	41.20	1.000	0.984	0.975		0.984	0.993	
65.28	25.70	0.973	0.981	0.993		0.915	0.963	
18.29	7.20	0.680	0.416	0.630		0.613	0.928	
0.03	0.01	0.424	0.122	0.376		0.288	0.886	
Maximum peak area		296,641	85,127	22,461				

(a) From bottom of fuel stack.

Table 7. Relative Axial Concentrations of Fission Products in Fuel Rod UN 100

Axial position (a)	Isotope, E_{γ} (keV)	^{137}Cs	^{134}Cs	^{106}Ru	^{95}Zr	R_{134}/R_{137}	R_{106}/R_{137}	R_{95}/R_{137}
		661.6	795.8	621.8	756.0			
cm	Inches	R_{137}	R_{134}	R_{106}	R_{95}			
267.08	105.15	0.437	0.172	0.454		0.394	1.038	
252.86	99.55	0.575	0.335	0.564		0.584	0.980	
212.22	83.55	0.846	0.816	0.903		0.965	1.068	
183.01	72.05	0.912	0.940	0.973		1.032	1.068	
158.37	62.35	0.960	1.000	0.953		1.041	0.992	
131.95	51.95	0.956	0.998	1.000		1.044	1.046	
105.54	41.55	1.000	0.913	0.979		0.913	0.979	
66.42	26.15	0.949	0.820	0.914		0.863	0.963	
19.69	7.75	0.688	0.420	0.668		0.610	0.971	
0.13	0.05	0.395	0.106	0.374		0.269	0.948	
Maximum peak area		288,095	80,697	20,135				

(a) From bottom of fuel stack.

Table 8. Atomic Composition of Mixed Oxide Fuel Samples

Isotope	Sample										
	97-B	98-A	98-B	99-A	99-B	100-A	100-B	101-A	101-B	102-A	102-B
U-238	1.00E+06	1.00E+06	1.00E+06	1.00E+06	1.00E+06	1.00E+06	1.00E+06	1.00E+06	1.00E+06	1.00E+06	1.00E+06
U-236	635	696	894	556	755	408	452	536	718	468	641
U-235	3841	3821	2268	4454	3038	5175	2750	4596	3197	4881	3844
U-234	65	81	108	68	71	44	42	52	49	48	33
Pu-242	842	855	1356	695	1082	557	616	650	1052	607	841
Pu-241	1379	1712	1572	1566	1508	1347	1002	1494	1544	1422	1376
Pu-240	4895	4910	4595	4762	4908	4783	3614	4817	5016	4841	4912
Pu-239	5442	6164	3959	7108	4672	8245	3990	7367	4992	8040	5478
Pu-238	190	306	338	230	265	165	138	217	313	177	20
Nd-150	229	299	287	164	378	120	137	214	249	145	255
Nd-148	414	525	505	278	522	205	244	278	417	240	404
Nd-146	676	900	869	456	950	339	402	479	704	408	655
Nd-145	734	884	865	479	788	367	435	391	714	413	674
Nd-144	1171	1441	1554	740	1591	547	692	740	1207	654	1102
Nd-143	951	1157	996	645	1010	506	656	633	894	566	880
Nd-142	39	277	32	58	488	43	122	365	121	121	141
Am-241	625	129	78	752	966	976	773	1034	327	1282	172
Np-237	2.00E+06	9.16E+05	2.36E+06	2.99E+06	8.68E+05	2.39E+06	2.56E+06	3.27E+06	2.66E+06	3.07E+06	7.81E+05

Figure 3. Gross Gamma and ^{137}Cs Activity
Scans of Fuel Rods UN 097, UN
098, and UN 099

Figure 4. Gross Gamma and ^{137}Cs Activity
Scans of Fuel Rods UN 100, UN
101, and UN 102

Figure 5. Scans of ^{95}Zr Activity in
Fuel Rods UN 098 and UN
101

Figure 6. Relative Activities of ^{137}Cs , ^{106}Ru and Activity Ratio $^{138}\text{Cs}/^{137}\text{Cs}$ for Rod UN 097

Figure 7. Relative Activities of ^{137}Cs , ^{106}Ru and Activity Ratio $^{134}\text{Cs}/^{137}\text{Cs}$ for Rod UN 102

Figure 8. Relative Activities of ^{137}Cs , ^{106}Ru and Activity Ratio $^{134}\text{Cs}/^{137}\text{Cs}$ for Rod UN 098

Figure 9. Relative Activities of ^{137}Cs , ^{106}Ru and Activity Ratio $^{134}\text{Cs}/^{137}\text{Cs}$ for Rod UN 101

Figure 10. Relative Activities of ^{137}Cs , ^{106}Ru and Activity Ratio $^{134}\text{Cs}/^{137}\text{Cs}$ for Rod UN 099

Figure 11. Relative Activities of ^{137}Cs , ^{106}Ru and Activity Ratio $^{134}\text{Cs}/^{137}\text{Cs}$ for Rod UN 100

Figure 12. Irradiation Histories and Growth Periods of Short-Lived Isotopes Detected

5. DISCUSSION

Gamma Scan Method

The gross gamma scans did not indicate any specific regions of high activity. This observation allows us to rule out axial migration of fission products since any such occurrence is expected to show up as localized activity, particularly at the interpellet gaps.

The interpellet gaps in the fuel rods are indicated by a decrease in gamma activity seen in Figures 3 through 5. From the regular spacing of the inverted spikes, it can be concluded that there were no major changes in the physical form of the pellets due to irradiation. The only exceptions observed were the two wider than usual interpellet gaps in the scan of rod UN 101. It can be seen from the repeat scans of the same regions on the following day (shown at the ends of the scan in Figure 4), that one of the wider gaps at the upper end of the rod disappeared. This was attributed to a settling of fuel pellets due to the handling of the rods. This was further supported by a slight change in the positions of interpellet gaps in the later scan of the same region. The reproducible wider gap at the bottom of the rod was not considered serious enough to warrant a detailed investigation. This was further supported by the observation of identical interpellet gaps in the original and repeat gamma scans. It was, therefore, concluded that this wider gap originated at the time the fuel rod was assembled.

The isotopes for which relative activity data, R values, are given in Tables 2 through 7, were chosen for their mode of formation and half-lives. ^{137}Cs , ^{106}Ru , and ^{95}Zr being primary products of fission, their R values represent fuel burnup profiles during part or all of the irradiation period. The activity of ^{137}Cs detected would be from fuel burnup during the entire irradiation because of its long half-life. The R values of ^{106}Ru and ^{95}Zr portray relative fuel burnup profiles during a part of the irradiation (see Figure 12) with ^{95}Zr revealing the relative profile of late-in-life fuel burnup.

The R values of ^{134}Cs and the R value ratio, R_{134}/R_{137} , represent the square of neutron flux and the neutron flux, respectively, averaged over the entire irradiation period because of the fact that all ^{134}Cs formed during that period was counted.

A consideration of Table 5, in light of the above general conclusions, reveals the fuel burnup profiles of fuel rod UN 101 during various stages of irradiation. The maximum fuel burnup for the entire irradiation of 8.33 years occurred, based on the R_{137} value, at about 106 cm from the bottom of the fuel stack. The values of R_{106} indicate that during the later 5-year period, maximum fuel burnup rate took place at about 159 cm while late-in-life R_{95} values show that the fuel burnup rate is higher at the ends of the rod. This is also reflected in the R value ratios, R_{106}/R_{137} , where the values greater than unity represent increased fuel burnup rate at later periods of irradiation compared to the average burnup during the entire irradiation.

The time averaged neutron flux, on the basis of the ratio R_{134}/R_{137} , and its squared value represented by R_{134} , show maximum values at the same axial position of about 159 cm.

The relative fuel burnup rates in other fuel rods and the respective neutron fluxes experienced by these rods during the entire irradiation, as well as portions of such irradiations, can be interpreted similarly with the aid of tables of R values and Figures 6 through 10. Analyses of this nature reveal that the maxima of both R values and their ratio with respect to R_{137} occur at different axial positions in different fuel rods. In addition, a change in these maximum activity positions occurred during the irradiation period. The former is attributable to the position of the rods in a fuel assembly and the location of the assembly containing the fuel rod within the core of a reactor. The latter, namely changes in the position of maximum fuel burnup rates during irradiation, is associated with the power drift along the fuel rod axis.

The absolute fuel burnup determined by radiochemical analysis of two transverse sections of each fuel rod represent values for the entire irradiation. Therefore, the relative axial concentration profiles of ^{137}Cs , R_{137} , are employed in evaluating conversion factors to construct absolute fuel burnup profiles. Since two sections of each rod were analyzed radiochemically, it is more appropriate to utilize an average of the conversion factors calculated for the

two axial positions in converting relative values to absolute fuel burnup profiles. These average values of conversion factors are given in Table 9.

The axial locations of samples that were analyzed by radiochemical methods are identified relative to the bottom of the fuel rods. The axial positions in determining relative fuel burnup profiles, on the other hand, are referenced relative to the bottom of the fuel pellet stack that was detected with ease in the gamma scans. Therefore, it is necessary to rationalize the two different reference positions. This was achieved with the help of the specification drawings of the fuel rod assemblies. It was determined from the available schematic that the bottom position of the fuel pellet stack is located 15.24 cm (6.0 in.) from the bottom of the fuel rods.

The relative atomic composition of irradiated mixed oxide fuel rods (Table 8) is based, as explained earlier, on mass and gamma spectroscopic data. The number of atoms of ^{238}U remaining in the fuel after irradiation is chosen as an internal reference. The atomic abundance of ^{237}Np based on the intensity of its characteristic gamma ray, 86.5 keV, appears to yield rather high values. All possible interferences with and sources of this gamma ray were considered but no reasonable explanation could be found. At the same time, it is not possible to identify the source of this isotope to justify its observed abundance in the fuel samples analyzed.

Table 9. Absolute Fuel Burnup and Conversion Factors for Mixed Oxide Fuel Rods

Sample number	Axial position ^(a)		Relative activity ^(b) of ^{137}Cs (R_{137})	Fuel burnup			Average fuel burnup at % per unit, R_{137}
	cm	Inches		Nd method	GWd/mt	Per unit rel. act.	
97-A	250.19	98.5	0.699	--	--	--	--
97-B	77.47	30.5	0.974	1.68	16.14	1.725	1.725
98-A	250.19	98.5	0.760	1.92	18.48	2.525	2.630±0.105
98-B	77.47	30.5	0.936	2.56	24.53	2.735	
99-A	247.65	97.5	0.740	1.47	14.10	1.986	2.22±0.234
99-B	80.01	31.5	0.970	2.38	22.82	2.454	
100-A	247.65	97.5	0.711	1.06	10.18	1.491	1.529±0.137
100-B	80.01	31.5	0.940	1.66	15.93	1.766	
101-A	247.65	97.5	0.773	1.40	13.49	1.811	1.951±0.140
101-B	80.01	31.5	0.914	1.91	18.38	1.090	
102-A	247.65	97.5	0.779	1.22	11.69	1.566	1.493±0.073
102-B	80.01	31.5	0.958	1.36	13.06	1.420	

(a) From bottom of fuel rod (Table 1).

(b) From Tables 2 through 7.

6. CONCLUSIONS

It was shown here that the R values of primary fission products represented the relative fuel burnup profiles. The burnup profiles during various segments of the irradiation period can be established by choosing a number of fission products with different half-lives, thus providing a method to unfold the overall burnup profile represented by the R value of a long-lived fission product such as ^{137}Cs .

The axial distribution of a neutron capture product, ^{134}Cs , was shown to represent the square of the time-averaged neutron flux while the ratio, R_{134}/R_{137} , signified the time-averaged neutron flux itself. This provides an avenue to determine the distribution of neutron fluxes along fuel rod axes for the entire irradiation period. Statistical analyses of this nature are worthwhile endeavors for obtaining information on neutron fluxes experienced by fuel assemblies and fuel elements.

The axial position of maximum R values used in the interpretation of results from the gamma scan technique was one of the 10 locations at which gamma spectra were recorded and analyzed. It is, therefore, conceivable that the true positions of maximum R values were not identified. This can be ascertained either by recording and analyzing gamma spectra at closer intervals or by fitting the observed pairs of activities and axial positions to an analytical function.

In the former approach, the precision of measurement is determined by the duration for which the gamma spectrum was stored during analysis as the fuel rod is scanned continuously at a uniform rate. However, adaptation of this technique should await the design and development of more reliable hardware and software. The latter method of establishing an analytical function of activity versus axial position based on a finite number of observations is easily adaptable. Even though this has not been done in the present investigation, results from similar work currently in progress at the Lynchburg Research Center appear to be promising.

7. REFERENCES

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