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PLUTONIUM UTILIZATION PROGRAM
TECHNICAL ACTIVITIES QUARTERLY REPORT
SEPTEMBER, OCTOBER, NOVEMBER 1969

January 1970

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PLUTONIUM UTILIZATION PROGRAM
TECHNICAL ACTIVITIES QUARTERLY REPORT
SEPTEMBER, OCTOBER, NOVEMBER 1969

By

Staff of Battelle-Northwest
Program Leader: F. G. Dawson

January 1970

OFFICE OF
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FOREWORD

The Plutonium Utilization Program is conducted by the Pacific Northwest Laboratory for the USAEC. The objective of the Technical Activities Quarterly Report is to inform the scientific community in a timely manner of the technical progress made on the program. The report contains brief technical discussions of accomplishments in all areas where significant progress has been made during the quarter. The results presented should be considered preliminary and do not constitute final publication of the work. A list of publications and papers is given in the report. Anyone wishing to obtain additional information on the work presented is encouraged to contact the author directly.

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September, October, November 1967	BNWL-654
December 1967, January, February 1968	BNWL-739
March, April, May 1968	BNWL-828
June, July, August 1968	BNWL-907
September, October, November 1968	BNWL-963
December 1968, January, February 1969	BNWL-1039
March, April, May 1969	BNWL-1105
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1.0 SUMMARY

REACTOR NEUTRONICS

A new version of the nonlinear least squares fitting program LEARN has been written. Both reliability and speed for difficult problems will be increased using the new version.

The analyses of experimental data collected in the AEC/PNL-CNEN cooperative program has been completed and a document describing the results has been drafted for publication.

Analytical Monte Carlo studies were made to determine the error expected in the analyses of the Batch Core resulting from assuming a one-dimensional model for a 19-rod cluster fuel element. The results of these calculations show that major errors in neutronic characteristics do not occur using a one-dimensional cell model.

The relative reaction rates of plutonium and uranium have been measured for rods containing a solid solution of $\text{UO}_2\text{-PuO}_2$ and a mixture of $350 \mu \text{ PuO}_2$ particles in UO_2 in the PCTR test cavity with a 4-in. graphite lattice. The measured data are in good agreement with the relative reaction rates calculated for the same fuels with the THERMOS-GRANIT Code which uses a special geometry routine to treat finite resonance shielding of particles embedded in cylindrical media by the method of Lane, Nordheim, and Sampson.

FUELS TECHNOLOGY

Detailed postirradiation examination of selected fuel rods from the Batch Core Experiment is being conducted to evaluate the irradiation performance of the different fuel

types. Axial gross gamma scans of irradiated rods containing cold-pressed-sintered and vibrationally compacted, pneumatically impacted $\text{UO}_2\text{-PuO}_2$ fuel show a relatively smooth distribution of fission product activity that is related to the axial power profile.

Detailed postirradiation evaluation of a variety of $\text{UO}_2\text{-PuO}_2$ fuels irradiated in PRTR fringe positions is in progress. The axial gross fission product distribution within the rods is indicative of the physical characteristics of the various fuel types. A rod containing cold-pressed-sintered $\text{UO}_2\text{-PuO}_2$ fuel was found to develop a defect in the cladding during removal from the element.

Postirradiation examination of four vibrationally compacted $\text{UO}_2\text{-PuO}_2$ fuel rods instrumented to monitor internal pressure during irradiation was initiated. Results of the gas sampling are in good agreement with calculated data. Fission gas and helium constitute over 99% of the total gas collected. Reduction of the free void volume in the rods during irradiation indicated the formation of a significant fraction of closed porosity within the fuel.

A comparison of the fuel structures formed in eight vibrationally compacted rods irradiated in the FERTF illustrates the rod-to-rod differences in low burnup fuels subjected to the same irradiation histories.

An assembly for defect testing different pellet fuel designs in the ETR M-3 loop has been designed. Heat transfer calculations indicate the maximum fuel temperatures for the different pellet fuel designs operating at 23 kW/ft are 2400 °C, 2570 °C, and 2100 °C for solid pellet-small diametral gap, solid pellet-large diametral gap, and cored pellet rods, respectively.

An experiment designed to monitor internal gas pressure in five PWR-sized mixed-oxide fuel rods during irradiation will be conducted in the ATR Water Loop under operating conditions typical of those projected for advanced PWR's.

A thermocouple instrumented experiment designed to monitor temperatures in mixed-oxide fuels during irradiation will be performed in the ETR M-3 loop.

The High Exposure Plutonium Studies program was reactivated with study emphasis to be placed upon the radiation doses associated with the fabrication of thermal recycle fuels and upon thermal reactor fuel geometries.

Studies have been initiated to investigate the dissolution properties of thermal reactor mixed-oxide fuels.

Studies have been initiated to evaluate the problems associated with the decontamination of reactor systems contaminated with ceramic plutonium-bearing fuel materials.

REACTOR SAFETY

The transient behavior of oxide fuels is being investigated by irradiations in SPERT and TREAT. A joint program involving BNW, General Electric, and Idaho Nuclear Corporation is being formulated to study the possible effects of adding PuO_2 particles to UO_2 fuels. Capsules are also being designed for studying the transient behavior of pelleted and vibrationally compacted fuels in the TREAT facility.

PRTR

The deactivation of PRTR was essentially completed during this report period. The PRTR equipment has been transferred into a "Laid-Up Equipment" category pending future reactivation or dismantling of the facility. The PRTR heavy water inventory has been returned to Savannah River. Seventy fuel elements will be stored at PRTR for possible reactivation of the facility. The remaining fuel material will be chemically reprocessed.

2.0 REACTOR NEUTRONICS

NONLINEAR LEAST-SQUARES DATA ANALYSES

D. A. Kottwitz

A new version of the nonlinear-least-squares fitting program LEARN,⁽¹⁾ called LEARN-I-B has been written and will eventually supercede LEARN-I-A.⁽²⁾ Two important changes have been made in the iterative portion of the program. The overall effect is to increase both reliability and speed for difficult problems. Reliability has been improved by requiring that each iterative step satisfy an accuracy criterion, i.e. a specified level of agreement between the predicted and actual consequences of a step. Speed has been increased by reversing the direction of a matrix step when it rises toward a "saddle" point. In addition, two new options involving the iterative procedure have been added. At execution time, the user may now elect (1) to allow a saddle solution as well as a minimum solution, and/or (2) to omit the calculation of the second partial derivatives in his THEORY subroutine. Debugging of the new version is almost complete.

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EXPERIMENT AND CALCULATED RESULT FOR UO_2 AND UO_2 - PuO_2
FUEL H_2O MODERATOR LOADINGS

L. D. Williams, J. B. Edgar, P. Loizzo*, R. Martinelli*
and N. Pacilio*

A cooperative program between USAEC and Italian Comitato Nazionale per L' Energia Nucleare (CNEN) was conducted at Argonne National Laboratory (ANL) and at Pacific Northwest Laboratory (PNL) under the auspices of the Plutonium Utilization Program. The purpose of this cooperative program was to provide experimental information on the neutronic characteristics of plutonium fuel in systems representing a typical Boiling Water Reactor (BWR). As a part of this program, plutonium fueled experiments were conducted at PNL in the Plutonium Recycle Critical Facility (PRCF). Uranium fueled experiments were included in the program for purposes of comparing core characteristics for uranium and plutonium enriched fueled systems. Interim results from the program have been published.⁽¹⁾ The analysis of all experimental data has been completed and a final report is being issued. A brief summary of the contents of the document is given in this report.

The loadings studied included both single zone (i.e., one fuel) and multizoned criticals. The effects of varying degrees of nonuniformity on core parameters were systematically investigated. The core parameters included:

- k_{eff}
- Kinetic Parameters (β_{eff}/λ and Rossi α)
- Flux and Power Distributions
- Moderator Temperature Coefficients
- Control Blade Worths

* CNEN Scientist

Power distributions were deduced from gross counting of fission products. Different fuels were intercalibrated by calorimetric measurements. Rossi alpha measurements were made in critical and subcritical loadings using the "endogenous" pulsed source method. (2)

Analytical correlations of some of these measured data were based primarily on CNEN developed theoretical models. (3) The reliability of the various models and the validity of the approximations used were checked by comparison with experiments and with other more sophisticated calculational schemes. The results of these correlations provide confidence to the analyses of studying plutonium recycle in the GARIGLIANO STATION in Italy.

An extensive analytical correlation of these experiments using the PNL developed methods is currently underway.

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ANALYTICAL STUDIES OF ONE AND TWO-DIMENSIONAL CELL
REPRESENTATIONS FOR CLUSTER GEOMETRY FUEL

D. H. Thomsen

This study was directed towards increasing the understanding of the neutronics of the Batch Core Experiment(1-4) in the Plutonium Recycle Test Reactor (PRTR). The PRTR fuel is in the form of 19-rod clusters. A two-dimensional calculation of the lattice cell is required to represent all of the rods correctly. However, most reactor physics cell codes are restricted to one dimension. The rods of the cluster are often represented in one dimension as annular fuel rings in the lattice cell calculation. This approximation has potential for introducing some error. The purpose of this study was to determine the magnitude of the errors incurred in using this one-dimensional cell approximation.

The method of solution was to compare the results of two calculations. One calculation represented the exact lattice configuration and the other a one-dimensional cell approximation. The cross sections and calculational methods in both calculations, using the Monte Carlo Code,⁽⁵⁾ were kept constant and only the cell geometry was different. The first calculation used a full two-dimensional representation of the 19-rod cluster and the cell boundary. The second calculation used a one-dimensional representation of the lattice cell.

Calculations were performed for a situation representing the Batch Core and for a natural UO_2 fueled cell. The setup representing Batch Core conditions was a cell containing a nonirradiated Batch Core element, i.e. UO_2 -2 wt% PuO_2 , while the moderator contained 11.25 ppm boron. The second calculational setup was for a natural UO_2 fuel element with unborated moderator. This fuel was chosen because of available experimental data and to allow deducing separately the effects due to uranium and plutonium.

The effects on the lattice cell constants of using the one-dimensional model are shown in Table 2.1. The difference quoted in this table is the one-dimensional value minus the two-dimensional value. These results show a relatively small effect on k_{∞} for both cells. Both values are within one standard deviation, and an upper limit would probably be ± 5 mk. Such a small difference for k_{∞} , however, is due to thermal and nonthermal effects compensating in the mixed oxide cell. The difference in the thermal effects (qf) are due predominately to the differences in thermal utilization. In addition, the differences in ηf is the only difference significantly outside of statistics.

TABLE 2.1. Comparison of Calculated Cell Parameters for One and Two Dimensional Models of the 19 Rod Cluster

Cell Parameter	Calculation	Difference, %
Multiplication (k_{∞})	$\text{PuO}_2\text{-UO}_2$	-0.002 ± 0.003
	UO_2	0.003 ± 0.004
Thermal Neutron Produced Absorption (qf)	$\text{PuO}_2\text{-UO}_2$	-0.005 ± 0.002
	UO_2	0.002 ± 0.002
Thermal Utilization (f)	$\text{PuO}_2\text{-UO}_2$	-0.004 ± 0.003
	UO_2	$+0.001 \pm 0.002$
Thermal to Total Absorption (p)	$\text{PuO}_2\text{-UO}_2$	0.001 ± 0.007
	UO_2	0.002 ± 0.004
Total Fission/Thermal Fission	$\text{PuO}_2\text{-UO}_2$	0.003 ± 0.011
	UO_2	0.000 ± 0.010

Parameters which should infer differences in initial burnup reaction rates are compared in Tables 2.2 and 2.3. The ^{238}U and ^{239}Pu capture rates appear to be calculated about 1% low for the UO_2 and $\text{UO}_2\text{-PuO}_2$ cases by the one-dimensional model. This difference would result in underpredicting both the production and destruction of ^{239}Pu so that the concentration during burnup may be fairly close. The error incurred in

the ^{238}U capture rate appears to be due to resonance absorption rates. The ^{235}U reaction rates are calculated very accurately by the one-dimensional model. The ^{240}Pu concentration would probably be underestimated as the ^{239}Pu capture is too low and the ^{240}Pu capture is too high. It is difficult to estimate, from these calculations, the effects on the ^{241}Pu and ^{242}Pu concentrations.

TABLE 2.2. Comparison of Burnup Parameters

<u>Parameter</u>	<u>Calculation</u>	<u>Difference, %</u>
^{238}U capture	$\text{PuO}_2\text{-UO}_2$	-1.2 ± 0.6
	UO_2	-0.8 ± 0.8
^{238}U Epithermal Capture	$\text{PuO}_2\text{-UO}_2$	-2.6 ± 1.0
	UO_2	-1.6 ± 1.6
^{235}U capture	$\text{PuO}_2\text{-UO}_2$	-0.5 ± 0.5
	UO_2	-0.2 ± 0.8
^{235}U Fission	$\text{PuO}_2\text{-UO}_2$	-0.16 ± 0.6
	UO_2	-0.2 ± 0.8

TABLE 2.3. Comparison of Burnup Parameters for the $\text{PuO}_2\text{-UO}_2$ Cell

<u>Parameter</u>	<u>Difference, %</u>
^{239}Pu capture	-1.1 ± 0.6
^{239}Pu Fission	-0.6 ± 1.0
^{240}Pu capture	1.9 ± 2.5
^{238}U capture	-1.2 ± 0.6
^{235}U capture	-0.5 ± 0.5

The relative effects on spatial reaction rates are shown in Table 2.4. The large difference for the UO_2 fuel case could well be misleading due to poor convergence.

TABLE 2.4. Comparison of Spatial Reaction Rate Distributions

	<u>Calculation</u>	<u>Difference, %</u>
Fission Per Rod (Outer Ring/Inner Ring)	$\text{PuO}_2\text{-UO}_2$	2 ± 1
	UO_2	0 ± 2
^{238}U Capture/Rod (Outer Ring/Inner Ring)	$\text{PuO}_2\text{-UO}_2$	1 ± 2
(Epithermal)	UO_2	11 ± 5

A comparison of calculated and experimental values for the lattice cell parameters for the natural UO_2 fuel cell are shown in Table 2.5. The ENDF/B cross sections were used for this calculation. The values in Table 2.5 are not defined in the same manner as are those in Table 2.1 but, rather, are calculated on the basis of the experimental definition. Note the close agreement between the two sets of calculated values. The disagreement in calculated and experimental values of k_∞ is believed to be real, while the discrepancy in ϵ may still be due to a lack of agreement in the definition. The cause of the discrepancy in k_∞ is not known. The agreement in all parameters except η^* leads to suspecting the thermal spectrum calculation or the cross sections for ^{235}U and/or ^{238}U .

TABLE 2.5. Comparison of Measured and Calculated Lattice Parameters

<u>Parameter</u>	<u>Experimental</u>	<u>Calculated</u>	
		<u>2-Dimensional</u>	<u>1-Dimensional</u>
k_∞	1.052 ± 0.003	1.040 ± 0.002	1.043 ± 0.003
ϵ	1.018 ± 0.002	1.024 ± 0.002	1.024 ± 0.002
p	0.868 ± 0.009	0.866 ± 0.002	0.867 ± 0.002
f	0.896 ± 0.003	0.899 ± 0.001	0.900 ± 0.001
η^*	1.329 ± 0.014	1.304 ± 0.006	1.305 ± 0.006

$$\eta^* = \frac{k_\infty}{\epsilon p f}$$

On the basis of this study, the errors introduced by using a one-dimensional model are concluded to be relatively small for both the situation representing the Batch Core Experiment and the UO_2 cell, and are probably overshadowed by errors in theory and/or cross sections. The magnitude of the error in the lattice cell constants would probably be less than 5 mk. The errors in burnup reaction rates would probably be in the order of 1%. The errors in local power distribution, i.e., within the cluster may, however, be emphasized during burnup.

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COMPARISON OF CALCULATED VERSUS MEASURED RELATIVE REACTION
RATES WITH RESPECT TO PARTICLE SIZE IN $\text{UO}_2\text{-PuO}_2$ FUEL RODS*

A. D. Vaughn

Introduction

Fuel element development for plutonium recycle has led to fuels composed of the mixed oxides of plutonium and uranium. The fabrication processes presently used in manufacturing of plutonium fuels result in distribution of the plutonium as particles throughout the uranium. In these fuels, power generation occurs mainly in the plutonium particles. Due to self-shielding properties of the particles, the reactivity of the fuel depends upon the particle size. Experimental and theoretical information about the effect of particle size on the characteristics of the reactor system is important for determining its impact on safety and economics.

Special fuel rods containing specific PuO_2 particle sizes were manufactured for a test lattice which could be inserted into the Physical Constants Testing Reactor (PCTR).⁽¹⁾ The experimental data necessary to determine the infinite medium neutron multiplication factor for particle size fuels in a graphite moderated lattice (4-in. pitch) have been obtained as a function of PuO_2 particle size ranging from a solid solution state to 350 μ . These determinations required the measurement of relative reaction rates of the fissionable isotopes with respect to particle size, and the relative reaction rates are presented in this report.

Plutonium and uranium fission foils were irradiated separately in specially constructed fuel rods. One set of foil holding rods was manufactured with solid solution $\text{UO}_2\text{-PuO}_2$ fuel, and another set with 350 μ particles of PuO_2 mixed with

* Portions of this work will be included in A. D. Vaughn's Master of Science Thesis Project in Nuclear Engineering to be submitted to the University of Washington

UO₂. Special foils were designed and fabricated to determine the self shielding of the particles in the 350 μ fuel rods. These foils contain 350 μ PuO₂ particles encased in aluminum.

All measurements were taken in the PCTR at room temperatures and the results are in excellent agreement with THERMOS-GRANIT Code^(2,3) calculations of the same parameters.

Discussion

Description of Measurement Facilities

An 8 × 8 array of 4-in. square, graphite lattice cells were stacked into the PCTR test cavity to provide the test assembly for these measurements. The four center cells of the array contained rods of either the solid solution set or the 350 μ set, depending on the measurement. Sixty cells surrounding the four center cells contained UO₂-2 wt% PuO₂ homogeneous fuel (Figure 2.1).

The reactor was made critical by the addition of PCTR drivers around the test cavity. The neutron energy spectrum and flux pattern were balanced across the test lattice by adjustment of the PCTR driver locations in the radial and axial directions, and by the addition of natural uranium and copper flux modifiers at the edge of the test lattice.

The match of the spectrum and the flux distributions were determined by irradiating bare and cadmium covered gold foils throughout the test lattice, and repositioning the flux modifiers until no significant energy or flux gradients existed across the cell boundaries around the four center test cells. This standard technique was used prior to measuring infinite medium neutron multiplication factors in the PCTR.

Description of the Particle Fuel Rods and Test Cells

Separate sets of four UO₂-2 wt% PuO₂ (8% ²⁴⁰Pu) fuel rods, one set containing plutonium-oxide in a solid solution state,

FIGURE 2.1. Fuel Arrangement in and Around PCTR Test Cavity

and another set containing 350 μ particles, were manufactured for the PCTR. These 0.566-in. OD rods were clad with 0.030-in. thick zirconium.

Two of the rods were built as separable units with a 0.005-in. thick cap at one end and a thin sleeve to contain

foils for irradiation in the rods in the center of the PCTR Test Cavity. Therefore, for each irradiation measurement, the four-rod center cell contained two complete fuel rods of a specific particle size and two sets of similar but separable particle rods containing foils at the axial center of the rods. The separable cells are depicted in Figure 2.2. However, the copper neutron absorbers at the cell boundary, depicted in the figure, were not present when those measurements were made.

Description of Task

Special plutonium foils were fabricated for the measurements in the 350 μ particle fuel rods. Aluminum discs (0.5-in. OD) were drilled with holes, each sufficiently small to contain a single 0.014-in. diameter PuO_2 particle. Each disc could contain approximately 45 particles. After insertion of the particles, the discs were clad in 0.005-in. thick aluminum covers. A diagram of the particle foils with respect to the particle rods is shown in Figure 2.3.

The plutonium reaction rate measurements in the solid solution rods were determined from a homogeneous foil of Al-1.8 wt% Pu, also encased in a 0.005-in. thick aluminum cover as depicted in Figure 2.4.

The ^{235}U reaction rates were determined from homogeneous foils of Al-7 wt% ^{235}U similar to the plutonium-aluminum foils but without the aluminum covers.

Theory of Measurement

Prior to the experiment, the fission foils used in this experiment were irradiated simultaneously on a graphite rotator in the PCTR thermal column.⁽⁴⁾ The flux in the thermal column was sufficiently constant so that any differences in count rate could be attributed to differences in the mass of fissile

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FIGURE 2.2. PCTR Center Test Cells with Separable Fuel Element and Foil Holders

FIGURE 2.3. Diagram of Particle Foils and Particle Fuel

FIGURE 2.4. Diagram of PuAl Foils and Solid Solution Fuel

material in the foils, provided the foils were of similar geometry and material. Therefore, the relative masses of the foils were determined by this method prior to the reaction rate measurements in the particle fuel.

The relative reaction rates of the particle foils with respect to the PuAl foils were also determined from this thermal column measurement. The results of the simultaneous irradiations of all the foils in a similar location were required by the analysis process. The measured parameters were as follows:

$$\frac{A_{\text{Fuel (ss)}}^{\text{PuAl}}}{A_{\text{Thermal Column}}^{\text{PuAl}}} - \text{Relative activity of PuAl foil in solid solution fuel rod to a similar foil irradiated simultaneously in the PCTR thermal column}$$

$$\frac{A_{\text{Fuel (350)}}^{350 \text{ Pu}}}{A_{\text{Thermal Column}}^{350 \text{ Pu}}} - \text{Relative activity of 350 } \mu \text{ particle foil irradiated in 350 } \mu \text{ particle fuel rod to a similar foil irradiated simultaneously in the PCTR thermal column.}$$

The differences due to the mass variations in these quantities could be eliminated by the data derived in the thermal column irradiations conducted prior to the experiment.

After correcting the measured data both for the residual counts taken before the irradiation and for background and flux peaking or dipping due to the presence of the foil (all these are mentioned later), essentially the following term was derived:

$$\frac{A_{\text{Fuel 350}}^{350 \text{ Pu}}}{A_{\text{Thermal Column}}^{\text{PuAl}}} - \text{Relative activity of 350 } \mu \text{ particle fuel in 350 } \mu \text{ particle rod to PuAl foil in thermal column}$$

This parameter cannot be directly measured because of the self-shielding differences due to the particle effect. From the following equation, one can see that the absolute mass of the foils must be known.

$$\frac{A_{\text{Fuel } 350}^{350 \text{ Pu}}}{A_{\text{Thermal Column}}^{\text{PuAl}}} - \frac{A_{\text{Fuel } 350}^{350 \text{ Pu}}}{A_{\text{Thermal Column}}^{350 \text{ Pu}}} \times \frac{A_{\text{Thermal Column}}^{350 \text{ Pu}}}{A_{\text{Thermal Column}}^{\text{PuAl}}} \frac{(\text{Mass PuAl in Thermal Column})}{(\text{Mass } 350 \mu \text{ foil in Thermal Column})}$$

The relative reaction rates of the ^{239}Pu isotope then could be determined by comparison with a similar parameter measured in the solid solution fuel rods, namely:

$$\frac{A_{\text{Fuel}}^{\text{PuAl}}}{A_{\text{Thermal Column}}^{\text{PuAl}}}$$

where the mass differences in this parameter can be directly removed by the previous and simultaneous irradiation of both foils in the thermal column.

It should be noted that the absolute weight of the fissionable isotopes in the foils must be known quite accurately in order to determine the relative reaction rates. Because these masses are on the order of 2 to 3 mg, most of the errors in the results were due to uncertainties in the masses.

Description of Measurements and Irradiations

Because only two rods in each set of four were separable, a plutonium foil was irradiated in one of the rods and the uranium and copper foils were irradiated together in the second rod but separated by a 0.001-in. thick aluminum catcher foil.

Identical foils were irradiated in the PCIR thermal column during each irradiation for normalization purposes.

The initial irradiations were done with bare foils, followed by irradiations with cadmium (0.040-in. thick) covering the foils in the separable rods. The foils in the PCTR thermal column were irradiated bare.

Each irradiation lasted 2 hr at a power level of 100 W. Counting started 8 hr after the irradiations so that the short-lived fission products had decayed to a negligible level. With only six foils per irradiation, the foils could be counted for a sufficient time and frequently enough to permit acquisition of excellent counting data.

The fission product decay gamma rays from the fission foils and the copper-64 decay gamma rays from the copper foils were counted using a sodium iodide detector and associated digital counting equipment. The energy discriminator was set to eliminate all counts from gamma rays with energy less than 600 keV.

Corrections to Foil Data

The unmodified relative foil activities were derived with respect to the foil activity in the thermal column by using the foil analysis code APDAC.⁽⁵⁾ This code corrects the data for background, subtracts residual counts derived by counting the fission foils before the irradiation, corrects for mass differences and decay time, and calculates the activation rates with respect to the thermal column foil with a statistical analysis of the results.

The data was exceptionally good even with the complicated decay rates associated with fission product activities. The error was less than 0.4% for the bare foils, and approximately 0.6% for the cadmium covered foils.

The results from the APDAC analysis needed further correction before use because of the peaking corrections in the fuel due to the presence of the foils, aluminum covers, and

end caps. Therefore, R - Z geometry transport theory calculations were performed for the foil and fuel diagrams depicted in Figures 2.3 and 2.4 using the DOT - SN⁽⁶⁾ Code. Cross sections for DOT were calculated with the THERMOS-GRANIT Code. The flux enhancement factor due to the presence of the foil in the fuel was obtained from the calculated ratio of the integrated flux in the foil area to a similar calculation using the same R - Z geometry, but with all fuel material in place of the foil and fuel. Thus, the correction factor was derived as follows:

$$\frac{A_o}{A_{\text{foil}}} = \frac{\phi_{\text{fuel}}}{\phi_{\text{foil}}}$$

where: A_o = The activity of the foil for flux in the area not affected by the presence of the foil
 A_{foil} = Measured foil activity
 ϕ_{fuel} = Integrated flux in foil region calculated by DOT - SN with fuel materials in all regions
 ϕ_{foil} = Integrated flux in foil region calculated by DOT - SN with foil and other materials as defined in Figures 2.3 and 2.4.

Similar R - Z calculations for the copper foils and the Al-7 wt% ²³⁵U foils irradiated in the fuel were also performed to obtain the flux enhancement factors in the fuel necessary to derive correct measurements of the reaction rates of the ²³⁵U and the nonfissionable isotopes of uranium and plutonium. These corrections amounted to approximately 2% of the unmodified-measured foil activities in the fuel.

THERMOS-GRANIT Calculations

The experimental lattice cells were mocked-up in cylindrical geometry for calculation using the THERMOS-GRANIT Code. This code employs a special geometry routine to treat finite resonance shielding of particles embedded in cylindrical media by the method of Lane, Nordheim, and Sampson.⁽⁷⁾ The relative

reaction rates of the fission isotopes ^{235}U and ^{239}Pu were calculated for five different particle sizes comprising solid solution, 50 μ , 100 μ , 200 μ , and 350 μ , respectively, to derive a functional description of the reaction rates with respect to particle size. To obtain this function, the calculated reaction rates for each isotope with respect to a specific particle size were normalized to the average flux in the fuel region. The differences in the average flux in the fuel region between each particle size case were corrected by a factor derived from the measured copper foil data.

It was assumed that the relative activities of the copper foils irradiated in the fuel would indicate the differences in the flux with respect to the particle size fuel rods and, therefore, would also be directly proportional to the correction factors necessary to normalize the calculated quantities for comparison purposes.

Measured Results

The results of the foil measurements are listed in Tables 2.6, 2.7, and 2.8, for the plutonium, uranium, and copper foils, respectively. The interpolated activities are also listed for similar foil types in the 50, 100, and 200 μ rods, assuming a self-shielding effect linear with respect to the particle size. The same parameters calculated with the THERMOS-GRANIT Code are almost linear, as depicted in Figures 2.5 and 2.6. These same quantities are currently being used in the derivation of the infinite medium neutron multiplication factors by the PCTR techniques.

TABLE 2.6. Relative Plutonium Foil Activities with Respect to PuO_2 Particle Size

	Solid Solution	Particle Size, μ			
		Interpolated Values			350
		50	100	200	
Sub Cadmium	10.37	10.09	9.81	9.26	8.42
	± 0.10				± 0.14
Epi Cadmium	0.564	0.559	0.555	0.546	0.533
	± 0.006				± 0.009
Total	10.93	10.65	10.37	9.80	8.95
	± 0.08				± 0.14

TABLE 2.7. Relative ^{235}U Foil Activities with Respect to PuO_2 Particle Size

	Solid Solution	Particle Size, μ			
		Interpolated Values			350
		50	100	200	
Sub Cadmium	7.64	7.69	7.74	7.84	7.99
	± 0.05				± 0.05
Epi Cadmium	0.666	0.665	0.664	0.662	0.658
	± 0.006				± 0.006
Total	8.30	8.35	8.40	8.50	8.65
	± 0.04				± 0.05

TABLE 2.8. Relative Copper Foil Activities with Respect to PuO_2 Particle Size

	Solid Solution	Particle Size, μ			
		Interpolated Values			350
		50	100	200	
Sub Cadmium	7.777	7.825	7.873	7.970	8.115
	± 0.037				± 0.039
Epi Cadmium	1.247	1.247	1.247	1.246	1.245
	± 0.010				± 0.010
Total	9.024	9.072	9.120	9.216	9.360
	± 0.036				± 0.038

FIGURE 2.5. Comparison of Calculated and Measured Self-Shielding Effects in Plutonium with Respect to Particle Size Fuel Rods (UO₂-PuO₂)

FIGURE 2.6. Comparison of Calculated and Measured Self-Shielding Effects in Uranium with Respect to Particle Size Fuel Rods of UO₂-PuO₂

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3.0 FUELS TECHNOLOGY

PRTR FUEL EVALUATIONS

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With the shutdown of the PRTR, the irradiation performance of the selected rods irradiated in the reactor is being evaluated by means of destructive postirradiation examination. In order to report the postirradiation evaluation results more logically, the PRTR fuel evaluations are divided into four groups as follows:

- High Power Density Fuel Evaluation
- Extended Burnup Fuel Evaluation
- Internal Pressure Monitoring Experiment Evaluation
- FERTF Fuel Rod Evaluation

The postirradiation examination status of each group is reported separately as follows:

High Power Density Fuel Evaluation

High Power Density design mixed-oxide fuels were irradiated in PRTR as part of the Batch Core Experiment (BCE) to peak burnups of approximately 11,000 MWd/MTM at power generations greater than those currently employed in commercial power reactors, i.e., actual peak linear power generations of 20 kW/ft with maximum fuel temperatures near melting. Although the majority of the 66 High Power Density 19-rod cluster fuel elements included in the BCE were vibrationally compacted powder fuel, pellet fuel fabricated by both the hot-pressed and the cold-pressed sintered processes was included.

Detailed postirradiation examination of selected rods from the BCE is being conducted to evaluate the irradiation performance of the different fuel types. A summary of the elements and rods from the BCE selected for detailed examination and the current status of the examination is presented in Table 3.1.

TABLE 3.1. Summary of PRTR High Power Density Fuel Rod Postirradiation Examination Status

Element Number	Rod No.	Fuel Type	Estimated Peak Burnup, MWd/MTM	Gamma Scan	Profilemeter Scan	Fission Gas Analysis	Burnup Analysis	Ceramography
6065	FE-74	UO ₂ -2 wt% PuO ₂ (VP)	5,000	X	Z	0	0	0
	FD-17	(VP)	6,500	X	Z	0	0	0
	FS-07	(VP)	10,000	X	Y	0	0	0
6519	FR-72	(VP)	5,700	X	Y	0	0	0
	FN-86	(VP)	3,700	X	Z	0	0	0
	FE-69	(VP)	2,800	X	Z	0	0	0
6520	FR-78	(VP)	11,000	X	Y	0	0	0
6700	A-14	(HP)	9,600	X	Y	0	0	0
	A-103	(CPS)	9,600	Z	Y	0	0	0
	A-102	(CPS)	9,600	X	Y	0	0	0
6701	A-23	(HP)	3,800	X	Y	0	0	0
	A-12	(HP)	3,800	Z	Y	0	0	0

(VP) = Vibrationally compacted, pneumatically impacted fuel rod.

(CPS) = Cold-pressed-sintered pellet fuel rod.

(HP) = Hot-pressed pellet fuel rod.

X = Examination completed

Y = Examination in progress

Z = Examination not scheduled

0 = Examination not started

Axial gross gamma scans were obtained on 10 of the 12 rods selected for detailed examination by Reactor Experiments Section personnel using a NaI detector. Fission product gamma-ray energy spectra were obtained from selected locations on the rods using a Ge(Li) detector. In general, the gamma scans of the hot-pressed $\text{UO}_2\text{-PuO}_2$ and the cold-pressed-sintered $\text{UO}_2\text{-PuO}_2$ pellet-containing rods are similar to the vibrationally compacted rods fabricated with pneumatically impacted $\text{UO}_2\text{-PuO}_2$ fuel material, and show a smooth distribution of fission product activity along the length of the rods that is directly related to the axial power profile (Figures 3.1 and 3.2). However, gamma scans of the higher burnup, higher power vibrationally compacted fuel rods indicate gross gamma activity discontinuities in the central regions of the rods which are probably associated with localized fission product migration or fuel movement (Figure 3.3). Postirradiation examination of these rods is continuing.

Extended Burnup Fuel Evaluation

A variety of experimental UO_2 and $\text{UO}_2\text{-PuO}_2$ elements containing 0.5, 1.0 and 2.0 wt% PuO_2 were irradiated in PRTR fringe positions to evaluate the irradiation performance of the various fuel types subjected to extended burnups. Detailed postirradiation examination of selected fuel rods irradiated in the PRTR fringe positions is in progress. The status of the examination of these rods is summarized in Table 3.2.

Full-length axial gross gamma scans were obtained on 13 of the 17 rods selected for detailed postirradiation examination by Reactor Experiments Section personnel using a NaI detector. Fission product gamma-ray energy spectra were also obtained from selected locations on the rods using a Ge(Li) detector.

Neg 0695526-3

FIGURE 3.1. Gross Gamma Scan of a PRTR Fuel Rod (A-102) Containing Cold-Pressed-Sintered UO_2 -1.94 wt% PbO_2 Pellet Fuel. Estimated peak burnup of 3,600 MWd/MTM.

Neg 0695526-2

FIGURE 3.2. Gross Gamma Scan of a Vibrationally Compacted PRTR Fuel Rod (FR-72) Containing Pneumatically Impacted, Vibrationally Compacted UO₂-2 wt% PuO₂ Fuel. Estimated peak burnup of 4,600 MWd/MTM.

Neg (0695526-1

FIGURE 3.3. Gross Gamma Scan of a Vibrationally Compacted PRTR Fuel Rod (FR-78) Containing Pneumatically Impacted, Vibrationally Compacted UO_2 -2 wt% PuO_2 Fuel. Estimated peak burnup 11,000 MWd/MTM.

TABLE 3.2. Summary of the Postirradiation Examination Status for Fuel Rods Irradiated to Extended Burnup in PRTR Fringe Positions

Element Number	Rod No.	Fuel Type	Estimated Peak Burnup, MWd/MTM	Gamma Scan	Profile-meter Scan	Fission Gas Analysis	Burnup Analysis	Ceramo-graphy
1037	A-1330	UO ₂ (SW)	14,500	X	Z	X	0	Y
	A-1377	↓	--	X	Z	X	0	Y
	A-1398	↓	--	X	Z	X	0	Y
	A-1325	↓	--	Z	Z	X	Z	Y
5118	DB-63	UO ₂ -0.5 wt% PuO ₂ (VP) (MM)	18,500	X	Z	X	Z	Y
	DA-48	↓	18,500	Z	Z	X	Z	Y
5186	ZZ-17	↓	--	X	Z	X	0	Y
	ZZ-7	↓	--	X	Z	X	0	Y
	zz-12	↓	12,500	X	Z	X	0	Y
	CN-5	↓	12,500	X	Z	Lost	Z	Y
	CN-7	↓	12,500	Z	Z	0	Z	Z
	CM-85	↓	12,500	Z	Z	0	Z	Z
5224	CS-27	UO ₂ -1 wt% PuO ₂ (SW) (PI)	13,500	X	Z	X	0	Y
5226	DF-80	↓	11,500	X	Z	X	0	Y
6003	5C-27	UO ₂ -2 wt% PuO ₂ (VP) (PI)	13,000	X	Z	0	0	0
6005	5F-36	↓	8,000	X	Z	0	0	0
6521	#6	ThO ₂ -5 wt% PuO ₂ (VP) (PI)	Low	X	Z	Lost	Y	Y

(VP) = Vibrationally compacted fuel rod

(SW) = Swage compacted fuel rod

(CPS) = Cold-pressed-sintered pellet fuel rod

(MM) = Heterogeneously enriched or incrementally loaded fuel material

(PI) = High-energy-rate pneumatically impacted fuel

X = Examination completed

Y = Examination in progress

Z = Examination not scheduled

0 = Examination not started

The axial fission product distribution within the rods is indicative of the different physical characteristics of the various fuel types and the irradiation conditions to which the rods were subjected. Activity discontinuities, indicative of localized fission product migration, occurred in the higher power regions of rods containing cold-pressed-sintered UO_2 - PuO_2 pellet fuel (Figure 3.4).

Vibrationally compacted UO_2 - PuO_2 fuel rods fabricated by incrementally loading mechanically mixed fuel showed random localized variations in fission product activity associated with plutonium nonuniformity (Figure 3.5). However, the randomness of the fission product distribution indicates that considerable mixing of the incrementally loaded UO_2 - PuO_2 fuel occurred during the vibrational compaction process. The scan of this particular rod (DB-63) also shows spikes which may be caused by localized fission product migration or localized fuel movement.

The axial fission product distribution in swage-compacted UO_2 - PuO_2 rods fabricated by incrementally loading mechanically mixed fuel shows pronounced activity variations corresponding to variations in plutonium concentration within the 1/2-in long increments (Figure 3.6). The swage-compaction process does not result in significant mixing of the incrementally loaded UO_2 - PuO_2 fuel material and the spatial plutonium distribution remains in the as-loaded condition.

Fuel Rod ZZ-12 from PRTR FE-5186 (Figure 3.4) developed a defect in the cladding while the rod was being removed from the element in the PRTR basin. The defect, located about 2 in. down from the top end plug weld, was evidenced by a stream of gas bubbles (Figure 3.7). A small crack appears to be associated with the defect although there is no evidence of localized clad swelling. A moderately heavy crud deposit was

FIGURE 3.4. Axial Gross Gamma Scan of a PRTR Fuel Element (ZZ-12) Containing Cold-Pressed-Sintered UO_2 -0.5 wt% PuO_2 as Fuel. Estimated peak burnup 12,500 MWd/MTM.

Neg 06955Z-1

FIGURE 3.5. Axi-1 Gross Gamma Scan of a Vibrationally Compacted PRTR Fuel Rod (DB-63) Fabricated by Incrementally Loading Mechanically Mixed UO_2 -0.5 wt% PuO_2 Fuel. Estimated peak burnup 18,500 MWd/MTM.

Neg 0695526-5

FIGURE 3.6. Axial Gross Gamma Scan of a Swage-Compacted PRTR Fuel Rod (CN-5) Fabricated by Incrementally Loading Mechanically Mixed UO_2 -0.5 wt% PuO_2 Fuel. Estimated peak burnup 12,500 MWd/MTM.

FIGURE 3.7. A Stream of Fission Gas Bubbles Emanating from a Defected Zircaloy-Clad PRTR Fuel Rod (ZZ-12) Containing Cold-Pressed-Sintered UO_2 -0.5 wt% PuO_2 Pellet Fuel. Peak burnup 12,500 MWd/MTM.

present in the defect region. It is assumed that an incipient failure existed in the cladding and that the vibration associated with rod removal caused complete penetration because there is no evidence of external physical damage and the gas bubbles indicate the presence of an internal pressure. Fuel Rod ZZ-12 was one of three rods in FE-5186 containing cold-pressed-sintered UO_2 -0.5 wt% PuO_2 pellets. The remainder of the 19 rods contained swage-compacted fuel material. The element, one of the first mixed-oxide elements loaded into PRTR, had an estimated peak burnup of 12,500 MWd/MTM. A detailed postirradiation examination will be conducted to determine the cause of the defect.

Fission gas analysis has been completed on 11 of the 17 rods included in the group of extended burnup fuel rods.

The composition of the gas collected from each rod is summarized in Table 3.3. The quantity of gas collected from Rod ZZ-12, considerably less than expected, is indicative of a defect. Analysis of the fission gas data is in progress.

Internal Pressure Monitoring Experiment Evaluation

Vibrationally compacted $\text{UO}_2\text{-PuO}_2$ fuel rods instrumented to monitor internal gas pressure during irradiation were successfully irradiated in PRIR to determine the effects of fuel temperature and burnup on both sorbed and fission gas release. The postirradiation examination status for the four instrumented rods is summarized in Table 3.4.

Because of problems encountered during the gas sampling operation, the gas from Rod No. 4 was not collected. The difficulties encountered were possibly indicative of a leak in the rod or the associated instrumentation hardware. However, neither problems during irradiation or physical damage was evidenced. Results of the postirradiation gas sampling from the two high burnup and one low burnup fuel rods are summarized in Table 3.5. The composition of the gas collected from each rod is summarized in Table 3.6.

The released fission gas (Xe + Kr) volume in the vibrationally compacted rods, as calculated from the measured rate of internal pressure increase during irradiation and the fractional release - volumetric average fuel temperature relationship developed previously, agree with the quantities obtained during postirradiation sampling. However, it is interesting to note that the measured postirradiation fuel void volume is only approximately 50% of the initial as-fabricated fuel void volume (assuming the gas plenum volume does not change significantly as a result of irradiation). This reduction in available fuel void volume is undoubtedly a result of changes in fuel morphology occurring during irradiation

TABLE 3.3. PRTR Extended Burnup Fuel Rod Fission Gas Analysis

Rod No.	Gas Volume, cm ³ (STP)	Molar Percent									
		Xe	Kr	CO ₂	Ar	O ₂	N ₂	CO	He	H ₂	CH ₄
A-1325	45.5	58.03	6.52	0.02	0.15	0.07	0.21	<0.10	35.0	<0.01	<0.01
A-1398	44.5	54.87	6.36	0.05	0.02	0.01	0.16	<0.10	38.6	<0.01	<0.01
A-1377	38.0	51.09	6.04	0.03	0.05	0.11	0.32	<0.10	42.4	<0.01	<0.01
A-1330	34.1	49.93	6.08	0.04	0.09	0.09	0.34	<0.10	43.4	<0.01	<0.01
DB-48	112	78.32	5.04	<0.01	0.16	0.03	0.10	<0.10	16.4	<0.01	<0.01
DB-63	113	72.41	4.68	0.01	<0.01	<0.01	<0.01	<0.10	22.8	<0.01	<0.01
CN-5	130	Sample Lost During Analysis									
CS-27	261	62.42	4.51	<0.01	<0.01	<0.01	<0.01	<0.10	33.1	<0.01	<0.01
DF-80	256	82.09	5.81	0.02	0.02	<0.01	0.10	<0.10	11.9	<0.01	<0.01
ZZ-17	16.1	7.08	0.53	0.01	0.01	0.04	0.14	<0.10	92.2	<0.01	<0.01
ZZ-7	22.5	6.82	0.59	0.02	0.03	0.14	0.92	<0.10	91.5	<0.01	<0.01
zz-12	24.4	49.25	3.94	0.05	0.48	0.43	41.6	<0.10	4.26	<0.01	<0.01

TABLE 3.4. Summary of PRIR Instrumented Fuel Rod Postirradiation Examination Status

Element Number	Rod. No.	Fuel Type	Estimated Peak Burnup, MWd/MTM	Gamma Scan	Profilometer Scan	Fission Gas Analysis	Burnup Analysis	Ceramo-graphy	Clad Metallography
6519	2	UO ₂ -2 wt% PuO ₂ (VP) ↓	6,000	Z	Z	X	Y	Y	Y
	4			Z	Z	Lost	Y	Y	Z
6520	7		11,000	Z	Z	X	Y	Y	Y
	8			Z	Z	X	Y	Y	Y

(VP) = Vibrationally compacted, pneumatically impacted fuel rod.

X = Examination completed

Y = Examination in progress

Z = Examination not scheduled

TABLE 3.5. Postirradiation Gas Sampling from Instrumented PRIR UO₂-PuO₂ Fuel Rods

Fuel Rod No.	Nominal Peak Power Generation, kW/ft	Peak Burnup, MWd/MTM	Calculated Released Fission Gas Volume,* cm ³	Measured Total Gas Volume, cm ³	Measured Fission Gas Volume, cm ³	Measured Helium Volume, cm ³	Measured Fuel Void Volume,** cm ³	As-Fabricated Fuel Void Volume, cm ³
2	9.5	6,000	22	45	23.5	21.4	11.1	22.5
7	16.5	11,000	110	145	114.5	30.5	13.0	22.5
8	16.5	11,000	110	149	117	32	--	22.5

* Based on the measured pressure increase within the rods during irradiation.

** Measured Fuel Void Volume = (Measured Rod Void Volume) - (Plenum Volume).

TABLE 3.6. Composition of Gas Collected from Instrumented PRTR $\text{UO}_2\text{-PuO}_2$ Fuel Rods

<u>Constituent</u>	<u>Rod No. 2, mole %</u>	<u>Rod No. 7, mole %</u>	<u>Rod No. 8, mole %</u>
Xe-136	19.7	30.7	30.4
-134	13.4	20.2	19.9
-133	<0.01	<0.01	<0.01
-132	9.74	14.0	14.0
-131	6.34	9.07	9.10
-130	0.02	0.05	0.03
Kr-86	1.43	2.41	2.43
-85	0.20	0.32	0.33
-84	0.85	1.43	1.40
-83	0.51	0.79	0.78
-82	<0.01	0.01	<0.01
-80	<0.01	<0.01	<0.01
-78	<0.01	<0.01	<0.01
CO_2	0.06	0.03	0.02
Ar	0.33	<0.01	<0.01
O_2	<0.01	<0.01	<0.01
N_2	0.10	0.08	0.11
CO	<0.10	<0.10	<0.10
He	47.4	21.0	21.5
H_2	<0.01	<0.01	<0.01
CH_4	<0.01	<0.01	<0.01

and represents the amount of closed porosity in the fuel, i.e. approximately $12 \text{ cm}^3/\text{rod}$. It is hypothesized that the major portion of the reduction in free void volume in the fuel, constituting approximately 25% of the rod void volume, occurred rapidly during the early stages of the irradiation inasmuch as the calculated rate of pressure increase throughout the irradiation (assuming a constant fuel void volume) agreed with the measured pressure buildup.

The amount of helium fill-gas collected from the rods during postirradiation gas sampling was less than the $\sim 45 \text{ cm}^5$ of preirradiated free void volume within the rods. This result indicates either the trapping of some of the helium in the closed porosity in the fuel, or that the loaded rod was not completely purged before welding the final end plug.

The composition of the gas collected from the irradiated rods is normal for vibrationally compacted, pneumatically impacted $\text{UO}_2\text{-PuO}_2$ fuel, and the quantity and composition of the gas collected from the two high burnup rods are nearly identical (Table 3.6). Fission gas (Xe + Kr) and helium constitute over 99% of the total gas collected, which confirms the observation made during the course of the experiment that released sorbed gases and moisture react rapidly with the fuel and/or cladding and do not contribute to the internal pressure within the rod.

FERTF Fuel Rod Evaluation

The objective of the fuel testing program in the PRTR Fuel Element Rupture Test Facility (FERTF) was to investigate the effects of specific power and burnup on the defect behavior of pellet and vibrationally compacted fuel rods. A specially designed 8-rod FERTF Test Element was utilized for these experiments.

The rod power generation in the FERTF was determined to within approximately $\pm 3\%$ by flow, differential temperature, and gamma scanning measurements. Relative power generation among rods in the 8-rod FERTF Test Element was determined by counting the gamma energy peaks associated with $^{95}\text{Zr}/^{95}\text{Nb}$ and $^{140}\text{Ba}/^{140}\text{La}$ with a NaI gamma spectrometer.

Eight identical vibrationally compacted $\text{UO}_2\text{-2 wt\% PuO}_2$ rods were irradiated together in the 8-rod FERTF Test Element

to study differences in rod-to-rod fuel structural features formed during irradiation under the same conditions. The eight rods studied were irradiated in the FERTF at a peak rod power generation of 16.4 kW/ft to a peak burnup of 380 MWd/MTM. For a coolant temperature of 255 °C, the calculated maximum core temperature was 2550 °C.

A low power, short duration irradiation of about 30 min (FERTF Test 9032) was performed with the eight new vibrationally compacted UO_2 -2 wt% PuO_2 rods at fuel temperatures (400 °C) below which fission product migration capable of producing misleading gamma scanning results could occur. Gamma scanning results of the activated rods showed that the ratio of peak-to-average power among the eight rods was approximately 1.01 (Figure 3.8). The identical assembly using the same fuel rods was subsequently irradiated at a peak power generation of 16.4 kW/ft (FERTF Test 9035) to compare the low and high power gamma scanning test results. After cooling periods of 28 days and 8 months, scanning results obtained from the eight irradiated rods confirmed that the rod-to-rod power variation was within +1% (Figure 3.8). The small rod-to-rod variations can probably be attributed to inherent differences in the vibrationally compacted rods and to statistical variations in gamma scanning.

Evaluation of the pre- and postirradiation profilometer data for six of the eight FERTF rods, irradiated under the same conditions for approximately two days, indicated no significant rod diameter changes as a result of irradiation. However, significant changes in the overall length were indicated (Table 3.7). Because there is no obvious explanation for the indicated length changes for rods irradiated under these conditions for such a short period of time, the validity of the postirradiation length measurements is subject to question and

FIGURE 3.8. Relative Power Generation Among Eight Vibrationally Compacted UO_2 -2 wt% PuO_2 Fuel Rods Irradiated in the PRTR Fuel Element Rupture Test Facility

TABLE 3.7. Profilometer Measurement Data on FERTF Rods

Rod No.	$\Delta D^{(1)}$ at 0° , mils	ΔD at 90° , mils	$\Delta H^{(2)}$ mils	AL, (3) mils	% AL
FS-41	≤ 1.0	≤ 1.0	≤ 1.0	-16	-0.02
FS-42	≤ 2.0	≤ 1.0	≤ 1.0	-15	-0.02
FS-43	≤ 1.5	≤ 1.5	≤ 0.5	-3	<-0.01
FS-45	≤ 2.5	≤ 2.5	≤ 1.0	-7	-0.01
FS-46	≤ 1.0	≤ 1.0	≤ 0.5	+38	+0.06
FS-48	≤ 2.5	≤ 2.0	≤ 1.0	+82	+0.12

(1) ΔD = Change in rod diameter.

(2) ΔH = Change in rod ovality

(3) AL = Change in rod length over a 67.3 in. measured length.

is being investigated further, although similar measurements repeated on nonirradiated rods agree to within ± 5 mils.

A comparison of the fuel structural features formed at the peak power position in the eight rods illustrates the rod-to-rod differences occurring in low burnup vibrationally compacted mixed-oxide rods subjected to the same irradiation histories (Figure 3.9). These rods were irradiated at a peak power generation of 16.4 kW/ft to a peak burnup of 380 MWd/MTM. Although the sintering boundaries among the eight rods as indicated by the beta-gamma and alpha-autoradiographs are comparable, there are differences in the sizes of the central voids and the morphology of the columnar grains formed during irradiation. The structural features of the fuel and the beta-gamma autoradiograph of one rod (FS-45) are indicative of fuel temperatures higher than those existing in the other seven rods. Evaluation of the data related to the relative performance of these eight FERTF rods is continuing.

FIG

FIGURE 3.9.

Transverse Sections from Eight Vibrationally Compacted UO_2 -2 wt% PuO_2 Fuel Rods with Identical Irradiation Histories. Transverse sections represent a linear rod power generation of 16.4 kW/ft and a burnup of 380 MWD/MTM.

DEFECT TESTING

M. D. Freshley and T. B. Burley

Irradiation tests of defected and nondefected mixed-oxide fuels will be performed under high performance conditions in the ETR M-3 loop facility. The defect testing program includes an investigation of three important parameters consisting of (1) fuel form (small and large gap solid pellet, dished pellet, and cored pellet), (2) power generation (23 of 27 kW/ft), and (3) burnup.

The defect test assembly will consist of 19 0.565-in. diameter rods similar to the PRTR fuel element design (Figure 3.10). Twelve fueled Zircaloy-clad rods, three of which can be replaced remotely, will be located in the outer 12-rod ring of the element. A flux wire assembly is located in the center support rod and the six-rod ring will be composed of dummy rods to balance the flow distribution within the element. A sleeve will surround the element to protect the pressure tube from possible damage during irradiation.

Different pellet fuel rod designs will be irradiated in the 12-rod ring of the assembly, and one of the replaceable rods will be defected for each experiment (Figure 3.10). Prefect irradiation of rods incorporating the different fuel designs will be conducted in the replaceable positions of the element. The mixed-oxide pellet rods include different designs for providing void space to accommodate fuel swelling and thermal expansion, i.e. different diametral gaps, dishing, and coring. The possible effects of large and small diametral gaps will be investigated. A dish volume of 3% will be used in the dished pellet design, and a core volume of 10% will be used in the cored pellet design.

Power generation calculations for the defect testing assembly with 12 fueled rods were performed by Reactor Experiment Section personnel using the THERMOS code. The results

ROD TYPE:

- 1 - SOLID (0.006" DIAMETRAL GAP)
- 2 - SOLID (0.010" DIAMETRAL GAP)
- 3 - DISHED (3% VOID VOLUME WITH 0.006" DIAMETRAL GAP)
- 4 - DISHED (3% VOID VOLUME WITH 0.010" DIAMETRAL GAP)
- 5 - CORED (10% VOID VOLUME)

FIGURE 3.10. Arrangement of the Different Types of Fuel Rods in the Defect Testing Assembly

indicate that a peak rod power generation of 22.5 kW/ft can be achieved in the ETR M-3 loop facility with rods containing UO_2 -4.0 wt% PuO_2 fuel (Figure 3.11). Concentrations of plutonium less than 4% produce undesirably low power generations, whereas those greater than 4% are nonprototypic and do not increase the power generation significantly.

CHEAT heat transfer calculations indicate the maximum fuel temperatures for the different pellet fuel rod designs operating at 23 kW/ft to be 2480, 2570, and 2100 °C for solid pellet-small diametral gap, solid pellet-large diametral gap, and cored pellet rods, respectively (Figure 3.12).

FIGURE 3.11. Calculated Peak Rod Power Generation Versus Plutonium Enrichment for Defect Testing Assembly Operating in ETR M-3 Loop Facility

FIGURE 3.12. Calculated Maximum Fuel Temperature Versus Rod Power Generation for Different Mixed-Oxide Pellet Fuel Designs Operating in Defect Testing Assembly in ETR M-3 Loop Facility

Fabrication of the defect testing element has commenced and thermal hydraulic flow tests have been completed on a mockup test assembly.

INSTRUMENTED FUEL TESTS

T. B. Burley and M. D. Freshley

Instrumented experiments to monitor internal gas pressure buildup and fuel temperatures in mixed-oxide fuel rods during irradiation will be conducted in the ATR Water Loop and the ETR M-3 Loop, respectively.

Internal Pressure Monitoring Experiment

An experiment designed to monitor internal gas pressure in five PWR-sized mixed-oxide fuel rods during irradiation will be conducted in the ATR Water Loop under operating conditions typical of those projected for advanced PWR's. Sorbed and fission gas release behavior will be monitored and compared for pellet-containing and vibrationally compacted fuel rods during irradiation to extended burnups of approximately 50,000 MWd/MTM. Some of the experimental parameters to be investigated are as follows:

- a Sorbed gas release behavior as affected by different cladding materials (ZR-2 or stainless steel).
- Fission gas release rate as affected by burnup rate. Fuel diameters and plutonium enrichments will be adjusted to yield similar rod power and fuel temperature conditions for different rod-to-rod burnup rates in the fuel.
- Fission gas release as affected by fuel form. Gas release from fuel rods containing solid, dished, and cored pellets, and vibrationally compacted fuel will be compared.

- Controlled power cycles will be initiated near the end of extended incremental burnup periods to investigate gas release mechanisms during shutdown-startup periods.

The experimental arrangement for the five rods in the element is shown in Figure 3.13.

Fuel Temperature Monitoring Experiment

A thermocouple instrumented experiment designed to monitor temperatures in mixed-oxide fuels during irradiation will be performed in the ETR M-3 Loop. Fuel temperatures will be monitored during a programmed power increase, during subsequent steady-state operation, and during a programmed power decrease to study and compare the thermal behavior and in-reactor grain growth kinetics of different fuel types.

The instrumented test element will consist of 19 0.565-in. diameter rods similar to the PRTR fuel element design, with

FIGURE 3.13. Arrangement of Different Types of Fuel Rods in Internal Pressure Monitoring Experiment Element

the 12 fueled experimental rods positioned in the outer 12-rod ring of the assembly. A flux wire assembly will be located in the center position and the six-rod ring will be composed of dummy rods to balance the flow distribution within the element. The arrangement of the different types of fuel rods in the fuel temperature monitoring experiment element is shown in Figure 3.14.

ROD TYPE

- 1 - SOLID (0.006" DIAMETRAL GAP)
- 2 - SOLID (0.010" DIAMETRAL GAP)
- 3 - VIPAC
- 4 - DISHED (3% VOID VOLUME)
- 5 - CORED (10% VOID VOLUME)

FIGURE 3.14. Arrangement of Different Types of Fuel Rods in Fuel Temperature Monitoring Experiment Element

A core volume of 10% will be used in the cored-pellet design, and the resulting reduced power generation will be compensated for by increasing the PuO_2 enrichment. A dish volume of 3% will be used in the dished pellet design. Standard pneuhatically impacted powder fuel will be used in the vibrationally compacted fuel rods. Tungsten-rhenium thermocouples will be utilized to monitor fuel temperatures in the six instrumented rods.

HIGH EXPOSURE PLUTONIUM STUDIES

R. C. Smith, L. G. Faust, and H. H. VanTuyl

The High Exposure Plutonium Studies program under PUP was reactivated with study emphasis to be placed upon the radiation doses associated with the fabrication of thermal recycle fuels and upon thermal reactor fuel geometries.

Two contracts with commercial mixed-oxide fuel suppliers for high burnup plutonium studies were initiated. One contract was negotiated with United Nuclear Corporation to make radiation measurements on BWR type recycled plutonium fuels containing approximately 18 wt% ^{240}Pu . A second contract was negotiated with General Electric Company to obtain spent fuel samples from the high burnup SA-1 fuel program. Gamma energy and isotopic composition measurements will be made on the plutonium obtained from the samples.

PLUTONIUM FUEL DISSOLUTION STUDIES

R. E. Lerch

The purpose of this activity is to study the dissolution properties of thermal reactor mixed-oxide fuels. For the foreseeable future, reprocessing of such fuels will be accomplished by the Purex process, an aqueous solvent extraction process. Successful application of the Purex process to thermal reactor mixed-oxide fuels demands knowledge of the

dissolubility (the fraction of plutonium and uranium dissolving when the mixed-oxide fuel is exposed to boiling nitric acid for a short period of time), and dissolution rate of such fuels in nitric acid as affected by fuel fabrication and irradiation history. This information is essential in evaluating the effectiveness of conventional head end technology, in aiding equipment and flow-sheet design, and for insuring that a critical mass of plutonium in the form of undissolved fuel residues will not accumulate in dissolver vessels.

Nonirradiated and irradiated UO_2 - PuO_2 fuel samples of various types from PRTR and other programs will be included in the study. Fuel samples are being classified according to fabrication history, PuO_2 content, and irradiation history for dissolubility and dissolution rate tests.

The dissolubility of the selected samples will be determined by boiling the fuel samples in 12M nitric acid for one 6-hr period, or two 6-hr periods if necessary. After the second 6-hr treatment, any residues will be dissolved in 12M HNO_3 -0.05M HF to determine residual uranium and plutonium content.

Dissolution rate studies will be conducted on samples showing complete or nearly complete dissolution in nitric acid alone. Rate studies will not be conducted on samples showing incomplete dissolubility because these samples would require treatment other than straight nitric acid dissolution (i.e., HNO_3 -HF mixture). The rate studies will emphasize the relative dissolution rates of plutonium and uranium. In order to investigate the effect of the different variables in a relatively short time, test fuel samples will be dissolved in a large excess (~ 100 moles H^+ /mole Pu) of concentrated (8 to 12M) nitric acid at reflux temperature.

The dissolution procedure will consist of heating the desired concentration and volume of nitric acid to a pre-determined temperature. A weighed pellet will then be added (time zero) and dissolution allowed to proceed for periods of up to 8 hr. Small (0.2 to 0.5 ml) samples of the dissolver solution will be extracted periodically. The experiments will be conducted at a relatively constant volume and acidity by adding nitric acid after each rate study sample has been removed. Dissolver sample solutions will be analyzed for both plutonium and uranium to determine whether preferential dissolution of either constituent occurs.

The necessary equipment for conducting both the solubility and dissolution rate experiments has been assembled and tested.

PLUTONIUM FUELS DECONTAMINATION STUDIES

J. R. Devine

Procedures normally used for uranium and uranium dioxide decontamination in reactor systems dissolve sintered plutonium oxides or oxides fired at high temperatures at undesirably slow rates. Use of fuel reprocessing solvents aggressive enough to dissolve plutonium oxides cause excessive corrosion of reactor structural components. These problems have prompted studies of the mechanisms and kinetics associated with the decontamination of reactor systems contaminated with ceramic plutonium-bearing compounds.

As part of this program, the dissolution of sintered UO_2 pellets has been studied to determine the relative importance of the rate of the surface reactions compared to the rate of the transport of reactants and products in the solvent. These results are then used to optimize reactor decontamination procedures and to clarify dissolution mechanisms.

From the data presently available, it has been tentatively concluded that the dissolution rate in 4N HNO_3 at 50 °C, passes from reaction control to diffusion control and then back to reaction control with increasing velocity of the fluid past the fuel pellet. In the 50 °C OPG solution (oxalate, peroxide, gluconate buffered acid), on the other hand, diffusion control is the predominant mechanism throughout the range of velocities tested, i.e. the same velocities as with the HNO_3 .

Recent dissolution tests performed on a new set of pellets yielded significantly different results than have previous tests. The reason is thought to be due to a crazed surface permitting the solution to penetrate the bulk material and increase the effective surface area. Experiments utilizing pellets with a smooth, solid surface will be repeated.

In addition to these experiments in which well defined surface areas can be determined, the dissolution rate of irradiated UO_2 - PuO_2 fuels will be measured even though the surface area of the irradiated fuel is poorly defined. The primary value of these tests will be in determining the dissolution behavior of different types of mixed-oxide fuels as affected by different irradiation conditions. Comparisons of the dissolution rate of the irradiated fuel with that of the unirradiated pellets may be possible by sizing the fuel particles or crushing unirradiated pellets.

4.0 REACTOR SAFETY

TRANSIENT TESTING

M. D. Freshley and T. B. Burley

A knowledge of the behavior of reactor fuels subjected to high energy short duration power excursions is essential to permit a complete appraisal of fuel performance and reactor safety. The transient behavior of oxide fuels is being investigated by irradiations in SPERT and TREAT to determine whether special considerations associated with adding PuO₂ enrichment to different types of UO₂ fuels would influence reactor operation.

A joint program involving Pacific Northwest Laboratory, General Electric Company, and Idaho Nuclear Corporation is being formulated to investigate the possible effects of adding PuO₂ enrichment to UO₂ fuels in different ways.

Another joint program involving Pacific Northwest Laboratory and Argonne National Laboratory was conducted to compare the transient behavior of pelleted and vibrationally compacted fuels by irradiations in the transparent vessel in TREAT. A few confirming experiments will be performed with pins designed to eliminate the possible effects of power peaking at the ends of the fuel column.

Two calibration capsules incorporating different amounts of hafnium absorber material at each end of the 5.5-in. long enriched UO₂ fuel column to suppress power peaking were irradiated in TREAT. Gamma activity measurements were made on the individual pellets following the low energy irradiations to determine the effect of the hafnium on the axial power profile (Figure 4.1). The results of increasing the hafnium

FIGURE 4.1. Measured Axial Power Profile in TREAT Calibration Capsules Containing Different Amounts of Hafnium Absorber Material at Each End of 5.5-in. Long Enriched UO_2 Fuel Column

thickness from 0.100-in. to 0.200-in. indicate a relatively small effect on the axial power profile and the persistence of power peaking at the ends of the fuel column.

THERMOS and Monte Carlo calculations were performed by Reactor Experiments personnel to determine a configuration of hafnium absorber material capable of providing the desired axial power profile. Results of the calculations indicate that a 0.050-in. thick hafnium sleeve surrounding each end of the capsule and extending to the enriched UO_2 -depleted UO_2 interface would reduce the power in the end regions to approximately 80% of the power at the midplane of the fuel column (Figure 4.2).

One additional pellet-containing calibration capsule, three pellet-containing irradiation capsules, and two irradiation capsules containing vibrationally compacted fuel will now be fabricated utilizing the modified design for completion of the test series in TREAT.

ENRICHED UO_2 FUEL
 (4.79% ^{235}U)
DEPLETED UO_2
HAFNIUM ABSORBER

FIGURE 4.2. Calculated Power Profile in TREAT Capsules Using a Modified Configuration of Hafnium Absorber Material

5.0 PLUTONIUM RECYCLE TEST REACTOR

K. L. Young

The deactivation of PRTR was essentially completed during this report period. The PRTR equipment has been transferred into a "Laid-Up Equipment" category pending future reactivation or dismantling of the facility. The PRTR heavy water inventory has been returned to Savannah River. Seventy fuel elements (2 wt% PuO_2 - UO_2) are stored at PRTR for possible reactivation of the facility.

The deactivation activity was documented and controlled by 50 specially prepared System Deactivation Procedures. In addition, 162 Equipment Deactivation Procedures were prepared to document modifications required for system deactivation, as well as lay-up of individual pieces of equipment. A total of 6 of the above procedures have yet to be completed. The unfinished procedures deal with the fuel element storage basin and will be done following removal of fuel material for chemical reprocessing.

The PRTR staff has been reduced from 57 people present at June 30, 1969, to a current level of 2. These remaining personnel will complete the required deactivation work, supervise the fuel shipment for chemical processing, and perform surveillance activities.

6.0 PUBLICATIONS AND PRESENTATIONS

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