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SHORT-TERM FAST FLUX EBR II IRRADIATION OF $\text{PuO}_2\text{-UO}_2$ FUEL PINS

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SHORT-TERM FAST FLUX (EBR-II)
IRRADIATION OF PuO_2 - UO_2 FUEL PINS

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ABSTRACT

Results of the initial fast flux irradiation of mixed oxide fuel pins in EBR-II are described. Major objectives of the test were to obtain data on short-term fuel performance in a fast spectrum and to serve as a precursor to high burnup irradiations. Six encapsulated fuel pins incorporating a variety

of design and fabrication parameters were irradiated to a relatively low burnup and examined. All of the specimens behaved satisfactorily, and specimens with target burnups of 50,000 to 100,000 MWd/Te were subsequently inserted in EBR-II.

SECTION I

INTRODUCTION

The $\text{PuO}_2\text{-UO}_2$ mixed oxide fuel is a prime candidate for fast power reactors. With demonstrated performance in thermal flux environments⁽¹⁾ and the availability of fast flux facilities, interest is turning toward mixed oxide fuel testing in a fast spectrum. Important fuel performance characteristics, such as fission-gas release, fuel-cladding interactions, and thermal performance, must be determined under conditions simulating as closely as possible those expected in a typical fast ceramic reactor.⁽²⁾

Low fuel cycle cost promises to be major potential advantage of fast reactors over other power reactor systems.⁽³⁾ If the lower costs are to be realized, high burnups (5 to 10 percent of heavy atoms) must be achieved. These burnups generally are higher than those which have been of interest for thermal reactors because there is considerably more penalty in the latter from the reactivity change caused

by fission product poisoning. Accordingly, a series of fast flux irradiations⁽⁴⁾ has been designed to demonstrate high burnup for mixed oxide fuel over a broad range of power levels. The short-term irradiation of six encapsulated fuel pins described in this report was conducted in EBR-II to obtain "proof-type" data prior to insertion of high burnup specimens. The specific objectives of the irradiation were:

- a. Gain low burnup experience in a fast flux.
- b. Examine the effects of a prototype neutron flux and temperature profile on fuel behavior, and compare the results with those obtained in thermal flux irradiations.
- c. Establish relationships between various fuel parameters and performance.
- d. Validate the fuel pin and capsule designs prior to the insertion of high burnup specimens in EBR-II.

SECTION II

SUMMARY

As the first in a series of fast flux irradiations, six encapsulated fuel pins contain-

ing $\text{PuO}_2\text{-UO}_2$ were irradiated in EBR-II. The irradiation period was 10 days (peak

burnup 1800 MWd/Te), and included one full power shutdown and recovery plus several smaller power cycles. Peak heat generation rates ranged from 14.8 to 16.8 kW/ft. The following fuel parameters were tested.

Fuel Density—85 to 95% T.D.

Stoichiometry—O/M 1.97, 2.00, 2.03

Fuel Compaction Process—Pellets and Vibratory Compacted

Fuel-to-Cladding Diametral Gap—1 to 6 mils

Axial Restraint

Cladding Material—Types 304 SS, 316 SS, 347 SS, and Incoloy 800

Fuel performance was determined by an extensive post-irradiation examination. The fuel pins were found to be in excellent condition. Slight diametral increases (less than 0.2 mil) were measured in the pellet rods at pellet interfaces. The vibratory compacted (vi-pac) pin did not exhibit this ridging effect.

Internal effects were related to the fuel temperature and thermal profile that resulted from the various fuel design conditions. Measured fission-gas release fractions ranged from 5 percent for the high density fuel to 28 percent for the low density, vibratory compacted powder, and generally increased with decreasing density. With the exception of one specimen, gas release increased with central void size.

For pellet fuel, the diametral gap typically decreased as a result of irradiation. The hyperstoichiometric fuel exhibited the largest change in gap size.

Central void formation and grain growth within the fuel occurred to varying degrees, but no axial fuel redistribution was evident. Evaluation of the macrostructures, with the heat ratings normalized, suggests that the size of the central void formed is primarily a function of fuel density and fuel-cladding diametral gap. The radius of grain growth decreases with decreasing stoichiometry. Radial fuel redistribution was least in hypostoichiometric fuel. As anticipated, the morphology of the irradiated mixed oxide is similar to that found in UO_2 tested in comparable power and temperature levels in a thermal spectrum.

No gross structural changes were evident in the cladding materials, which operated at calculated peak temperatures of up to 1070°F. A slight reaction zone and/or preferential sensitization was observed on the inner surface of several of the specimens. Such localized zones have been observed previously, with little or no correlation to burnup or duration of exposure.

Based on the satisfactory behavior of these specimens, high burnup subassemblies were inserted into EBR-II

SECTION III

EXPERIMENTAL DESIGN

3.1 FUEL PIN DESIGN

All of the 0.250-inch diameter $\times$ 35-1/2-inch-long fuel pins contained 20 wt % PuO_2 -80 wt % UO_2 (mixed oxide) fuel, with a natural- UO_2 upper blanket and fission gas

reservoir integral within the cladding. The fuel length was fixed at 14-7/32 inches to match the EBR-II core height.* Fuel pin

* The height of the core has since been reduced to 13-1/2 inches.

TABLE 3-1

FUEL DESIGN PARAMETERS^(a)

<u>Fuel Pin</u>	<u>Fuel Compaction Process</u>	<u>Average Stoichiometry (O/M)</u>	<u>Average Fuel Density (% TD)</u>	<u>Smeared Density (% TD)</u>	<u>Average Diametral Gap (inch)</u>	<u>Axial Gap (inch)</u>	<u>Cladding Material</u>
F1A	Pellets	1.998	95.3	93	0.0039	0.49	347 SS
F1B	Pellets	2.000	96.2	95	0.0025	0.48	347 SS
F1C	Pellets	2.030	95.6	94	0.0022	0.002	Incoloy 800
F1D	Pellets	1.978	95.3	93	0.0030	0.48	Incoloy 800
F1E	Pellets	1.984	89.2	87	0.0030	0.50	316 SS
F1F	Vi-pac	1.986	85.3 ^(b)	85	N.A.	0.31	Incoloy 800

(a) Coprecipitated 20 wt % PuO₂—80 wt % UO₂.
Cladding dimensions (nominal)—0.250-inch od × 0.015-inch wall.

(b) Smeared density.

designs for pellet and powder fuel are illustrated in Figures 3-1 and 3-2, respectively. As indicated, four of the pellet pins contained a coil extensometer to accommodate axial thermal expansion. The remaining pellet pin F1C, was axially restrained. The powder fuel pin contained stainless steel wool to hold the material in place during handling and provide space for expansion during operation. Pertinent fuel parameters are summarized in Table 3-1.

The mixed oxide powder was made by the coprecipitation of plutonium hydroxide and ammonium diuranate from a solution of uranium and plutonium nitrates. To obtain the desired power levels, it was necessary to enrich fully the uranium in the mixed oxide. The powder was formed into low density pellets, or vibratorily compacted as

prescribed. Details of the fuel fabrication processes are furnished in Appendix A.

3.2 CAPSULE DESIGN

The detailed capsule design is shown in Figure 3-3. Encapsulation was required so that secondary containment was provided in the event of a cladding failure. The external configuration of the capsule was fixed to conform with the subassembly design. The sodium annulus between the fuel pin and Type 304 stainless steel capsule wall served as a heat transfer bond. Each fuel pin was wrapped on a spiral pitch with a stainless steel spacer wire to minimize bowing. The sodium level was established at between 3-1/2 and 4 inches above the fuel. Sodium filling of specimens (F1A, F1B, F1C, and F1D) proved

FIGURE 3-1. PELLET FUEL PIN ASSEMBLY DRAWING

REV D 455

FIGURE 3-2. POWDER FUEL PIN ASSEMBLY DRAWING

[illegible]

- 13- FABRICATE CAPSULE PER SPEC. 21-A-4935.
 - 12- SODIUM LEVEL LIMITS FOR G19-G24, +2%, -1/4 FROM REF LINE SHOWN.
 - 11- FINAL LENGTH TO BE DETERMINED AT ASSEMBLY.
 - 10- FUSION WELDS ON CAPSULE O.D. TO BE SMOOTH AND PASS THROUGH A .750 $\pm$.005 ID. 1/2" WIDE RING GAGE
 - 9- PART #6 TO BE 4B/8" FINAL LG. TO BE DETERMINED AT ASSEM.
 - 8- DEGREASE ALL PARTS COMPLETELY.
 - 7- NUMBER IDENTIFICATION TO RUN CONSECUTIVELY, STARTING WITH F-I-A, F-I-B, F-I-C, ETC.
 - 6- EACH CAPSULE MUST BE IDENTIFIED ON THE CYLINDRICAL SURFACE OF THE TOP OF THE CAP, MARKING TO BE ALONG AXIS OF THE CAP ON TWO SIDES, 180° APART.
 - 5- FILL ALL VOIDS WITH REACTOR GRADE HELIUM.
 - 4- DIM. INCLUDES ALLOWANCE FOR WELD SHRINKAGE.
 - 3- ANGULARITY OF END PLUG AXIS, WITH RESPECT TO TUBE AXIS MUST NOT EXCEED .015".
 - 2- SCRAATCHES, NICKS, ETC. INTRODUCED IN FABRICATION
a) HANDLING SHALL NOT EXCEED .001" IN DEPTH.
 - 1- T.I.R. OVER CAPSULE LENGTH .005
- NOTES: 1

- 16 ASTM-A 276-60T-TP-304 $A' = .150$ $A'' = \frac{1}{8}$ $C' = \frac{3}{16}$
- 18 ASTM-A 276-60T-TP-304 $A' = .150$ $A'' = \frac{1}{8}$ $C' = \frac{1}{8}$
- 4 TP-304 STN 6TL PART TO BE FURNISHED B AND PER DWG NO EB-1-38449-A AND ALTERED AS SHOWN ABOVE.

NOTED	NAME	DATE
ADV. ENG'R.	W. K. K. K.	OCT. 9-'63
RML	R. W. K. K.	OCT. 11-'63

FIGURE 3-3. CAPSULE ASSEMBLY DRAWING

to be a formidable problem. Detection of voids in the lower slot and the annulus necessitated extensive reworking (shaking and centrifugation) of these capsules before the entrapped bubbles could be removed. This treatment caused some fuel cracking and relocation. The bonding problem subsequently was solved by narrowing the slot and filling under vacuum; capsules F1E and F1F were filled in this fashion. Details of the capsule fabrication are given in Appendix B.

3.3 SUBASSEMBLY DESIGN

The test vehicle was a standard EBR-II Mark A experimental subassembly which is designed to accommodate 19 cylindrical 3/8-inch diameter $\times$ 40-inch long capsules. In the Mark A design, the individual capsules are contained in internally dimpled shroud tubes. The 19 shroud tubes are tangent within the hexagonal subassembly tube; the spaces between the adjacent shroud tubes are filled with stainless steel strips to prevent coolant flow bypassing the capsules. This annular design provides uniform cool-

ant flow to each channel and prevents bowing of the capsules. The subassembly contains an internal orifice to provide the requisite coolant flow. Preliminary flow tests were conducted at ANL to select the proper orifice size (0.469-inch diameter).

Final flow testing on the completed subassembly was performed in a water loop at ANL, and the results converted to equivalent sodium flow. The 10 capsule channels divided the 20.5 gpm full flow equally because of their uniform hydraulic characteristics.

The test subassembly, identified as XG01, contained the six fuel capsules, two FCR materials capsules, and two Nuclear Superheat (PA-13)* materials capsules. The nine remaining spaces were filled with stainless steel rods. Flow channels for the dummy rods were plugged to ensure that each capsule would be cooled adequately if the reactor were operated at proportional flow and power.

* Results obtained in this experiment are to be described in a forthcoming publication.

SECTION IV

OPERATIONAL HISTORY

4.1 CAPSULE POSITIONS

The six fuel capsules were positioned in the subassembly according to Figure 4-1, which also shows the orientation of the subassembly in the reactor. The subassembly was supposed to have been oriented in reverse of that shown, but it developed subsequently that the index point on the EBR-II fuel handling gripper mechanism had been programmed 180 degrees out-of-phase.

4.2 REACTOR EXPOSURE

The subassembly was charged into the EBR-II storage basket on April 24, 1965, and inserted in reactor grid position 4-F-4 on May 6. The reactor achieved significant power on May 12 and was shut down on May 22. Figure 4-2 is a plot of reactor power versus time for the cycle. The integrated reactor exposure was 381 MWd at a maximum power level of 45 MWt. Full power was

FIGURE 4-1. ORIENTATION OF GROUP 1 CAPSULES IN EBR-II SUBASSEMBLY

FIGURE 4-2. INTEGRATED EXPOSURE DURING PERIOD: 381 MWd, MAY 1965

maintained during most of this period. The reactor sustained one full power shutdown and recovery plus several short power reductions. The startup procedure included a series of small oscillation tests.* The subassembly was discharged to the storage basket on May 23. The normalized axial flux profile for EBR-II is shown in Figure 4-3. Because of the absence of poisons in the reactor, the chopped-cosine shape and peak vertical position remained essentially constant during the irradiation. The radial flux gradient across the subassembly in position 4-F-2 was about 12 percent; the midplane fission rates in that position at 45 MWt were estimated⁽⁵⁾ as in Table 4-1. For these fission rates and the relative positions within the subassembly, linear power levels (at the beginning of the irradiation) were calculated for each of the specimens and are tabulated in Table 4-2. The estimated maximum neutron dose to the cladding was 1.2×10^{21} neutrons/cm² (>1MeV).

The sodium coolant inlet temperature and flow to the subassembly were maintained at

* Special trapezoidal and stepwise reactor stability studies.

700°F and 100 percent capacity, respectively, during operation. The individual flow past each capsule was thus held at approximately 2 gpm. Figure 4-4 includes a plot of the calculated coolant temperature rise through the shroud tubes.

4.3 DISCHARGE AND DISASSEMBLY

The subassembly was discharged from the tank storage basket on May 26 and transferred to the Fuel Cycle Facility (FCF) air cell for disassembly. During this procedure all surfaces were steam cleaned to remove the sodium. Disassembly was accomplished without difficulty by cutting the hex tube.

The fuel capsules appeared to have a somewhat darker surface than the materials capsules. The individual capsules were placed in two aluminum transfer baskets and stored overnight in the FCF. A thermocouple reading the following morning indicated a temperature of 113°F in the baskets at the midheight of the fueled section of the capsules. The baskets were then loaded in the EBR-II Source Coffin and transferred to Test Area North (TAN), where they were stored in the water canal for 25 days until the activity was low enough to permit shipment to the Radioactive Materials Laboratory at Vallecitos for detailed examination.

TABLE 4-1

RADIAL FISSION RATES AT REACTOR MIDPLANE FOR EBR-II POSITION 4-F-2

Isotope	Fissions/gm-sec		
	Maximum Pin	Center Pin	Minimum Pin
Pu-239	9.6×10^{12}	9.0×10^{12}	8.5×10^{12}
U-235	8.0×10^{12}	7.5×10^{12}	7.0×10^{12}
U-238	0.5×10^{12}	0.5×10^{12}	0.4×10^{12}

FIGURE 4-3. LOCAL TO AVERAGE CORE AXIAL POWER DENSITY RATIOS

FIGURE 4-4. PLOT OF TEMPERATURES VERSUS LENGTH BASED ON 15 kW/ft PEAK POWER

TABLE 4-2

PEAK HEAT GENERATION RATES BASED ON FISSION RATE DATA*

<u>Fuel Pin</u>	<u>Linear Power (kW/ft)</u>	<u>Peak Cladding Temperature (Inside Diameter) (°F)</u>
F1A	16.1	1086
F1B	16.4	1091
F1C	16.8	1097
F1D	16.2	1088
F1E	14.8	1066
F1F	16.4	1091

* Based on 192 MeV per fission.

SECTION V

POST-IRRADIATION EXAMINATION

5.1 GAMMA SCANNING

The fuel capsules were scanned axially to determine gross gamma, Ru-103 (0.5 MeV), and Ba-140 (1.6 MeV) activity profiles. The scans were obtained using conventional equipment by moving the vertically suspended capsule past a collimating slit and directing the beam at a NaI scintillation detector. A

discriminator was used to record the various gamma energy levels.

The results of the scanning are summarized in Table 5-1; all of the scans are noted in Appendix C. The gross scans ranged in appearance from the relatively smooth cosine-shaped profile exhibited by F1E and F1F, to the irregular contour displayed by F1C. This

TABLE 5-1

GAMMA SCANNING RESULTS

<u>Capsule Number</u>	<u>Gross Gamma</u>	<u>Ba-140</u>	<u>Ru-103</u>
F1A	Uniform scan except for possible fuel movement approximately 2 to 3-1/2 inches from the bottom of the fuel column.	Very similar to the gross gamma activity scan. The activity decreases are slightly less pronounced.	Some indication of Ru-103 migration toward the ends of the fuel column.
F1B	Extremely uniform scan except for two activity spikes located near the center of the fuel column. No fuel movement was indicated.	Similar to the gross gamma activity scan although a slightly less uniform profile was observed.	Nearly identical to the Ba-140 activity scan.
F1C	Very nonuniform scan indicating either gross fission product or fuel movement.	General activity profile the same as the gross gamma scan although the peaks were not as pronounced.	Same general profile as the gross gamma scan with less pronounced peaks and valleys.
F1D	Several pronounced peaks indicating the possibility of slight axial fuel movement.	Nonuniformity of the activity profile less pronounced than on the gross scan.	Similar to the Ba-140 scan.
F1E	Completely uniform activity profile.	Uniform scan.	Uniform scan.
F1F	Occasional small valleys in an otherwise uniform profile. The dips are probably the result of the powder compaction rather than fuel or fission product movement.	Uniform scan.	Uniform scan.

variation is typified by Figure 5-1. Multi-channel analysis of the peaks observed in F1C, as well as the single channel scans, indicated that some localized radial fuel relocation had occurred. When the fuel pins subsequently were sectioned, it was found that the irregularities in the scans could be correlated with cracks and gaps in the fuel that evidently were introduced during centrifugation and shaking to remove the bubbles.

5.2 CAPSULE DISASSEMBLY

Each of the sodium containers was pierced and a sample of the helium fill gas was analyzed for K4-85 to determine the integrity of the fuel pins. No activity was detected ($<10^{-4}$ percent) in any of the capsules, confirming that all of the pins were intact. The sodium container was cut circumferentially at the top and bottom end. The fuel pins were removed from the sodium-fill tubes by heating under a nitrogen atmosphere to approximately 350°F in a vertical heating jacket to melt the sodium. The fuel pin claddings were free of any cracks, blemishes, scratches, or other unusual markings which would indicate possible cladding damage.

5.3 DIMENSIONAL MEASUREMENTS

Length measurements were performed using a specially constructed dial gauge and surface plate. Comparison of the pre- and post-irradiation values is shown in Table 5-2. It is believed that the changes are due to bowing, or the relief thereof, and could have resulted from the pre-irradiation thermal treatment.

Pre-irradiation diametral measurements were made at 1-inch intervals with a micrometer. Post-irradiation measurements were performed with an in-cell contour profilometer capable of measuring variations of less than ± 0.1 mil in cylindrical samples. In addition, micrometer

measurements were made at 1 inch and 3 inches from the bottom of each pin to cross-check the profilometer measurements. No significant diametral changes (<1 mil) were detected, although the profilometer traces revealed slight ridging (less than 0.2 mil) at some pellet interfaces for fuel-cladding diametral gaps less than about 4 mils. The vi-pac pin (F1F) did not exhibit this effect. Representative traces for a pellet rod and the vi-pac rod are shown in Figure 5-2; all of the traces are presented in Appendix D along with the tabulated diameter measurements.

5.4 FISSION GAS COLLECTION AND ANALYSIS

The fuel pins were punctured in an evacuated system and the gases collected in a system of known volume to determine the total release. Aliquots were taken for Kr-85 gamma counting, gas chromatography, and mass spectrometry. Fission gas data obtained are tabulated in Appendix E.

The composition of the gas was determined by chromatographic analysis. The virtual absence of air in the samples indicated that they were not contaminated during collection. Mass spectrometric analyses were obtained on samples from F1E and F1F to measure the isotopic distribution of the krypton and xenon in the fission gas.

The Kr-85 release was determined by gamma spectrometry (counting photons from decay) and compared with the theoretical quantity generated (calculated from the burnup) to obtain the fractional release. The results are presented in Table 5-3; release fractions ranged from 5 percent for the high density fuel to approximately 28 percent for the low density vibratory compacted powder. A plot of the data, Figure 5-3, shows that the release generally increased with decreasing fuel density. With the exception of F1D,

FIGURE 5-1. EXTREME CONTOURS OF GROSS GAMMA ACTIVITY PROFILES FOR GROUP 1 CAPSULES

FIGURE 5—2. PROFILOMETER TRACES COMPARING POWDER AND PELLETED FUEL PINS

TABLE 5-2

LENGTH MEASUREMENTS

<u>Capsule</u>	<u>Length, inches</u>		<u>ΔL</u>
	<u>Pre-Irradiation</u>	<u>Post-Irradiation</u>	
F1A	35.493	35.485	-0.013
F1B	35.548	35.540	-0.008
F1C	35.574	35.576	+0.002
F1D	35.542	35.535	-0.007
F1E	35.575	35.558	-0.017
F1F	34.894	34.881	-0.013

TABLE 5-3

FISSION GAS RELEASE DATA FOR GROUP 1 FUEL PINS

<u>Fuel Pin</u>	<u>Density (% TD)</u>	<u>Relative Power</u>	<u>Center Void Radius (mils)</u>	<u>% Fission Gas Released*</u>
F1A	95.3	0.955	9.0	11.1
F1B	96.2	0.975	0.5	5.1
F1C	95.6	1.000	8.5	6.7
F1D	95.3	0.960	0	7.9
F1E	89.2	0.870	15	26.0
F1F	85.3	0.975	22	27.6

* Based on K-85 release; assumed yield 0.21%

gas release increased with central void size; and, with the exception of F1C, gas release increased with grain growth.

5.5 FUEL BURNUP ANALYSIS

A burnup sample was taken from near the midfuel length of each pin and the burnup was determined by radiochemical analysis of selected fission products. The low burnup precluded a supplementary determination by

mass spectrometry because the changes in heavy element isotopic composition were too small. The results, which are summarized in Table 5-4, differed with predicted values as calculated from published⁽⁵⁾ EBR-II fission rates. Since the respective errors for the measured and calculated burnups are estimated to be 15 and 10 percent, most of the individual values could fall within the experimental error; however, it is not obvious that the errors should be biased

FIGURE 5-3. FISSION GAS RELEASE VERSUS FUEL DENSITY FOR SHORT-TERM FAST FLUX IRRADIATION

rather than normally distributed. The burnup values and fission rates used in this report are 10 percent below those as determined from the published values. A more accurate assessment of this problem awaits analysis of the higher burnup specimens currently in-plate.

5.6 SECTIONING PROFILES

Each fuel pin was sectioned at selected positions along its length and the macrostructure examined. The sectioning diagrams and resulting axial profiles are shown in Figures 5-4 through 5-6. In general, the structural changes (central void formation and grain growth) are observed to follow the calculated axial fuel temperature profile. Restructuring of the fuel occurred to varying degrees in all of the pins, but no significant axial fuel relocation was evident. The structures were compared with the pre-

irradiation fuel stoichiometry, fuel densities, and fuel-cladding diametral gaps. Since the fuel pins operated at different power levels, to compare the specimens it is necessary to normalize the heat ratings. This can be done by calculating the $\int_{T_s}^{T_r} KdT$, where T_r is the radius at which a particular structural change occurs. The results of this analysis (Table 5-5) suggest that the size of the central void formed primarily is a function of fuel density and diametral gap, with a lesser dependence on stoichiometry. The radial changes are plotted against the various fuel parameters in Figures 5-7, 5-8, and 5-9. A notable example of these functional relationships is the relatively large center void found 4 inches above the bottom of the fuel column in pin F1A. The fuel in this region contained a relatively large gap and was of relatively low density, which accounts for the greater restructuring.

TABLE 5-4

GROUP 1 BURNUP RESULTS

<u>Fuel Pin</u>	<u>Peak Burnup (% of Heavy Element)</u>	
	<u>Calculated^(a)</u>	<u>Measured^(b)</u>
F1A	0.202	0.182
F1B	0.202	0.168
F1C	0.209	0.158
F1D	0.202	0.166
F1E	0.196	0.168
F1F	0.223	0.178

(a) Based on EBR-II fission rate data—175 MeV/f

(b) Weighted mean of Nd-148, Cs-137, Ce-144, and Zr-95 analyses.

TABLE 5-5

HEAT RATING TO CAUSE STRUCTURAL
CHANGES IN GROUP 1 FUEL PINS^(a)

<u>Fuel Pin</u>	<u>Radii of Structural Changes (mils)</u>			$\int_{T_s}^{T_{CGG}} KdT$	$\int_{T_s}^{T_c} KdT$
	<u>Equiaxed Grain Growth</u>	<u>Columnar Grain Growth</u>	<u>Central Void</u>		
F1A	92	60	9.0	26.8	38.1
F1B	82	50	0.5	30.9	38.9
F1C	110	75	8.5	20.9	38.9
F1D	80	40	0	34.0 ^(b)	>39.2
F1E	98	70	15	20.6	33.8
F1F	110	75	22	20.6	37.1

(a) T_{CGG} = Temperature at which columnar grain growth occurs (~1800°C).

T_c = Center temperature.

T_s = Fuel surface temperature.

(b) Coarse equiaxed grains.

FIGURE 5-4. SECTIONING PROFILE

FIGURE 5-5. SECTIONING PROFILE

F1E O/M 1.984

GEAP-5570

F1F O/M 1.986
(VIBRATORY COMPACTED)

FIGURE 5-6. SECTIONING PROFILE

FIGURE 5-7. RESTRUCTURING AS A FUNCTION OF FUEL DENSITY

FIGURE 5-8. RESTRUCTURING VERSUS OXYGEN-TO-METAL RATIO

FIGURE 5-9. RESTRUCTURING AS A FUNCTION OF DIAMETRAL GAP

5.7 METALLOGRAPHIC EXAMINATION

Several metallographic sections, longitudinal as well as transverse, were examined from each specimen. Alpha and beta-gamma autoradiographs were also prepared from select specimens. The results of the examinations are summarized below.

Figure 5-10 compares the fuel microstructure of three stoichiometries having a similar density and irradiated at equivalent power levels. The diametral composites were made at the peak power section of the fuel column. It is readily observed that the restructuring effects are progressively less with decreasing stoichiometry. The radii of equiaxed and columnar grain growth were measured and plotted against the fuel parameters (see Figures 5-7 through 5-9). Grain

growth appears to be more dependent on stoichiometry and diametral gap than on density.

The one anomaly noted in the irradiated microstructure was the ring of second phase (black phase) noted near the periphery in F1D. This structure was not observed in the as-polished condition but was resolved after etching. This structure is noted in Figures 5-10, 5-11, and 5-13. High magnification of the structure, Figure 5-11, indicates that the region appears to be a multi-phase structure and its location within the fuel follows definite temperature contours. Microcores of this second phase were examined by both electron microprobe and micro focus x-ray diffraction techniques. Both of these studies showed no difference between the bulk mixed oxide and the second phase.

BLACK PHASE

NOTE APPARENT
PORE MOVEMENT
DOWN THE THERMAL GRADIENT

O/M - 1.97 (F1D)

100X

NOTE APPARENT
PORE MOVEMENT
UP THE THERMAL GRADIENT

O/M - 2.00 (F1B)

100X

REDUCED TO 55 PERCENT
FOR REPRODUCTION

O/M - 2.03 (F1C)

100X

FIGURE 5—10. EFFECT OF STOICHIOMETRY ON MICRO-
STRUCTURE OF MIXED OXIDE FUEL
IRRADIATED AT COMPARABLE POWER
LEVELS

GEAP-5570

ETCHED

500X

FIGURE 5-11. COMPOSITE SHOWING DARK RING OBSERVED
AT HIGH MAGNIFICATION

100X

POWDER

100X

PELLET

REDUCED TO 55 PERCENT
FOR REPRODUCTION

FIGURE 5—12. MICROSTRUCTURE OF LOW DENSITY
FUEL IRRADIATED FOR SHORT TIME
IN EBR-II

FIGURE 5-13. COMPOSITE SHOWING BLACK PHASE AND AXIAL FUEL
MOVEMENT IN SPECIMEN FID

No variation in the U/Pu ratio was observed and it is suggested that the effect is due to a compositional variation in oxygen. The lack of this phase in other hypostoichiometric specimens in this and other studies is unexplained.

Microstructures from the low density pellet and vi-pac pins are compared in Figure 5-12. Although the pins had run at high central temperatures, the periphery of the fuel was still largely nonsintered and porous. Both of these specimens contained hypostoichiometric fuel, yet no dark ring was observed as in F1D. The grosser structural changes in the vi-pac fuel stem apparently from either operating at a 10 percent higher power than the low density pellet pin or a slightly lower effective conductivity in the powder fuel.

Figure 5-13 shows a longitudinal section of pin F1D, where plastic fuel flow occurred between two pellets. The space between the two pellets is assumed to have existed prior to irradiation and the downward fuel movement (creep) occurred during irradiation.

An interesting observation in all of the specimens was the distinct appearance of spherical pores in the equiaxed-columnar grain transition region. Figure 5-14a shows a typical photograph of these pores. Although it generally is considered that these pores move up the thermal gradient, there is evidence (see Figure 5-10, Pin F1D) that some of these voids were moving in the opposite direction (determined by trails behind the pores). The pores noted in Figure 5-14a can be observed to be migrating to grain boundaries. Figure 5-14b shows the center section of pin F1C where the porosity is coalescing at the fuel center line. This section is presumed to be the initial stage in the formation of a central void.

White particles and a gray phase were occasionally observed in the irradiated fuel. These phases were not in any great quantity and in general the irradiated mixed oxide fuel appeared to be typical of what one would observe in irradiated UO_2 . Figure 5-15 shows the fuel cross section of pin F1A. The subgrain structure typically noted in as-fabricated coprecipitated mixed oxide fuel can readily be noted in the etched fuel near the cladding periphery. Figure 5-16 shows etched sections from each fuel pin cladding in this series which indicate that no gross structural changes occurred in the cladding which operated at a peak temperature of 1070°F . No discontinuities in cladding structure were evident in areas adjacent to pellet interfaces.

5.8 FUEL AUTORADIOGRAPHY

Autoradiographs were taken from selected metallographic sections to determine the distributions of alpha and beta-gamma activity. Previous work has shown (6,7) that these are useful tools for complementing metallographic examination in interpreting fuel and fission-product location and behavior. Cellulose nitrate was used to record the alpha tracks. Beta-gamma exposures were made with emulsions on Kodak high resolution plates. It should be noted that the alpha tracking is predominantly a surface effect, whereas the beta-gamma rays are more penetrating. Typical autoradiographs are presented in Figure 5-17.

No significant variations in alpha activity were evident in the autoradiographs. And no fuel movement across the fuel-blanket interface was observed.

The beta-gamma radiographs showed a typical pattern for oxide fuel irradiated at high specific power. In all cases, high

ETCHED

(a) PORE MIGRATION TOWARDS FUEL CENTER

500X

AS POLISHED

(b) FORMATION OF CENTRAL VOID

50X

FUEL
CENTER
LINE

FIGURE 5-14. RADIAL POROSITY MIGRATION IN OXIDE FUEL

100X

500X

500X

500X

REDUCED TO 55 PERCENT
FOR REPRODUCTION

FIGURE 5—15. PEAK POWER CROSS SECTION
OF FUEL PIN F1A

F1A - 347

F1B - 347

F1C I-800

F1D I-800

F1E 316

F1F I-800

FIGURE 5-16. POST-IRRADIATION CLADDING CROSS SECTIONS FROM GROUP 1 PINS

BETA-GAMMA RADIOGRAPH

PHOTO MACROGRAPH

ALPHA RADIOGRAPH

GEAP-5570

FIGURE 5-17. TYPICAL AUTORADIOGRAPHS IN IRRADIATED MIXED OXIDE FUEL (FROM PIN F1E)

activity was observed at pellet interfaces, gaps, and in cracks. The dense columnar grains typically were slightly less active, occasionally surrounded by a band of lower activity corresponding with the grain size transition zone in the microstructure. The outer region of the fuel generally was equivalent to the columnar grain region in activity,

suggesting that the volatile fission products had migrated to and condensed in the cooler areas. Figure 5-18 shows a comparison of pre- and post-irradiation diametral gap sizes in the fuel pins. With one notable exception (F1B) all of the pins exhibited a decrease in diametral gap during irradiation.

SECTION VI

DISCUSSION OF RESULTS

6.1 FUEL STRUCTURAL CHANGES

As expected, the morphology of the fast flux-irradiated fuel was similar to that found in $\text{PuO}_2\text{-UO}_2$ irradiated in a thermal spectrum. A difference of opinion exists in the literature^(8,9) concerning the effect of flux spectrum on the radial temperature profile across a fuel pin; the Group 1 results substantiate Edwards' assertion that the effect is minor for comparable heat ratings.* There is no evidence from the Group 1 irradiation that the temperature at which structural changes occur is influenced by the flux spectrum.

The size of the central void was found to be more a function of fuel smeared density (pellet density and diametral gap) than stoichiometry. The effect of stoichiometry on the fuel microstructure is quickly noted in Figure 5-10. A comparison of the 2.00 and 2.03 stoichiometry specimens indicate that the 2.03 specimen operated at a significantly higher centerline temperature, as evidenced by the central void and equiaxed and columnar grain growth radii. On the same basis the 1.97 specimen exhibited an even cooler structure with massive grain growth in the central region. The large massive grains

observed in the 1.97 fuel were apparently formed by grain boundary migration rather than lenticular pore sweeping. Grain boundary migration in ceramic fuels is assumed to occur under thermal gradients lower than that required for the sweeping of lenticular shaped pores. On this basis the thermal gradient (center temperature) of the 1.97 fuel would be significantly lower than either the 2.00 or 2.03 fuel. This indirect comparison of the three stoichiometries suggest that under the test conditions in this experiment the effective thermal conductivity of 20 wt% PuO_2 mixed oxide fuels decreases with increasing oxygen content from 1.97 to 2.03. This conclusion of an apparent higher conductivity for hypostoichiometric mixed oxides is partially supported by earlier studies.^(10,11) The effect of stoichiometry on structure development observed in this experiment agree with NUMEC thermal flux irradiations on mixed oxide fuels.⁽¹²⁾ Since other mechanisms such as grain boundary migration contribute to grain growth⁽¹³⁾ it may be inferred that stoichiometry affects diffusion coefficients and activation energies as well as thermal conductivity.

It was observed that the diametral gap size decreased as a result of irradiation

* There is a gross effect on burnup profile.

FIGURE 5-18. COMPARISON OF PRE- AND POST-IRRADIATION DIAMETRAL GAPS

with the exception of fuel pins F1B and F1F. The latter contained powder fuel, and the 2-mil gap observed after irradiation is attributed to sintering and densification of the fuel. The increased gap in F1B is an anomaly, which might be explained by a fortuitous choice of the fuel section examined. The relatively larger decrease in gap size for F1C appears to be related to its high oxygen-to-metal ratio. Measurements of the plastic deformation of UO_2 have shown^(14, 15) that plasticity increases with increasing O/M ratio, and the mixed oxide would be expected to behave similarly.

An interesting observation in all of the specimens was the distinct appearance of moving pores, but perhaps even more noteworthy is the fact that they did not all move up the thermal gradient, as is commonly implied. As noted in Figure 5-14, there is apparently a strong driving force which moves the voids toward grain boundaries and in some cases these voids were noted to move perpendicular to the thermal gradient to reach a boundary.

6.2 FUEL THERMAL PERFORMANCE

Thermal conductivity in porous bodies varies with fractional porosity P according to the general expression:

$$k_p = k(1 - \alpha P)$$

where k is the thermal conductivity of 100 percent dense oxide, α is a constant, and k_p is the conductivity of the porous oxide. A reasonably good fit to the data was obtained for $\alpha = 2.6$, which is similar to the relationship found for UO_2 by Notley and MacEwan.⁽¹⁶⁾ Recently, a model was formulated⁽¹⁷⁾ in which α increases with increasing volume fraction of porosity. This model also yields good agreement between the observed and calculated temperatures.

6.3 FISSION GAS RELEASE

Post-irradiation punctures of a number of UO_2 fuel rods operated at high heat ratings have given release values which corresponded well with the maximum fuel rod volumetric average temperature⁽¹⁹⁾ or with the value

of the $\int_{T_s}^{T_c} KdT$.⁽²⁰⁾ Other investigators⁽²¹⁾

have obtained a correlation between release and the cross-sectional fuel area operating above 1800°C for UO_2 -1.5 percent PuO_2 powder fuel. In the subject EBR-II irradiation, essentially all of the above relationships pertain to the data. It appears, however, that a better correlation exists between fractional release and fuel operating above 1800°C than between release and a mean fuel temperature. The dependence of release on fuel density apparently is associated with the resulting temperature profile in the fuel, (i.e., higher fuel temperatures occurred in the lower density fuel).

6.4 FUEL-CLADDING MECHANICAL INTERACTIONS

It was observed that pellet fuel pins exhibited small circumferential ridges at pellet interfaces, whereas the pin containing vi-pac fuel showed no such cladding deformation. Ridging at pellet interfaces also has been found⁽²²⁾ in fuel rods irradiated in water reactors. Ridge formation in the EBR-II pins appeared to occur for initial fuel-cladding diametral gaps up to 4 mils. but ridge heights varied considerably along the length. It has been postulated that differential thermal expansion between the inner and outer portion of the pellets resulted in hourglassing of the pellets, which is transmitted into larger strains at pellet interfaces. The significance of the ridges in terms of operating life remains to be determined.

6.5 FUEL-CLADDING INTERFACE REACTIONS

Localized reaction products and/or sensitization on the inner surface of the cladding,

such as found in several of the Group 1 fuel pins, have been observed previously⁽²³⁾ in FCR irradiations. Little or no correlation has been found with burnup or duration of exposure.

SECTION VII

CONCLUSIONS

1. The irradiated mixed oxide fuel pins were in excellent condition. Fuel pin and capsule performance were as predicted. Based on satisfactory behavior of these low exposure specimens, high burnup subassemblies were inserted into EBR-II.
2. The fuel pins did not exhibit significant deformation even with axial restraint of the fuel column. Slight ridging of the cladding is associated with pellet interfaces and small initial diametral gaps.
3. Fission gas release varies inversely with fuel density.
4. No significant axial fuel redistribution occurred in the mixed oxide fuel operated at center temperatures near 2400°C.
5. Morphology of fast flux irradiated fuel is similar to that observed from thermal flux irradiations.
6. Fuel restructuring increased with decreasing smeared density.
7. The smeared thermal conductivity of the mixed oxide fuel was noted to decrease with increasing oxygen content based on structural changes.
8. No significant differences were observed in the behavior of the several austenitic alloy cladding materials tested.

APPENDIX A

FUEL FABRICATION

A.1 FUEL SYNTHESIS

All of the fuel was synthesized by the coprecipitation of plutonium hydroxide and ammonium diuranate (ADU). A flow diagram of this process is shown in Figure A-1. Plutonium and uranium nitrates were mixed with nitric acid and precipitated with ammonium hydroxide. Following precipitation, the material was washed, filtered, and then dried. The filtered cake was then reduced at 750°C in an atmosphere of 6 percent hydrogen-94 percent helium to give (Pu, U)O₂.

A.2 FUEL PREPARATION AND FABRICATION

The final preparation of the fuel is dependent on the compaction process to be used. Both pressing and sintering, and vibratory compaction were used in the Group 1 specimens.

A.2.1 Pelleted Fuel

The reduced powder initially was ball milled and screened. Only material less than 200 mesh was carried through for pellet fabrication. The screened material was pressed at low pressure (5 to 20 ksi) to densify it and screened again. Pellets were pressed in a double action die at pressures up to 80 ksi without a binder. The pellets were then sintered in a 6 percent hydrogen-94 percent helium atmosphere for 4 hours at approximately 1550°C. Figure A-2 is a flow diagram of the pelletizing process.

A.2.2 Vibratory Compacted Fuel

Powder for vibratory compacted fuel rods was prepared by crushing pressed and sintered pellets. Typical particle size fractions used in the process are given below.

TABLE A-1

GRANULATION SIZES FOR VIBRATORY COMPACTION

<u>Mesh</u>	<u>Weight %</u>
-6 + 10	65
-35 + 65	20
-200	15

The blended powder was loaded into the rod and vibratory compacted to the desired density. Following compaction, the rod was filled with an inert gas, welded and inspected.

Figure A-3 is a detailed flowsheet of the fuel pin fabrication process.

FIGURE A-1. FLOW DIAGRAM — PLUTONIUM HYDROXIDE AMMONIUM DIURANATE COPRECIPITATION PROCESS

FIGURE A-2. FLOW DIAGRAM — PRESSING AND SINTERING OF CERAMIC OXIDE PELLETS

FIGURE A-3. FUEL PIN FABRICATION FLOW SHEET

APPENDIX B

CAPSULE FABRICATION

The capsule fabrication flowsheet is given as Figure B-1. The encapsulation technique consisted of assembling the capsule parts around the fuel specimen and then filling the capsule with sodium to a prescribed level. The sodium-filled system, shown in Figures B-2, B-3, and B-4, provides sodium of known purity and permits thermal cycling of the completed capsules at 1050°F to ensure good thermal bonds. The system enables three capsules to be filled simultaneously. A dummy capsule containing 3/16-inch long $\times$ 1/16-inch wide nichrome parts to simulate voids in the sodium was used as the standard for the subsequent eddy current testing of finished capsules for sodium voids.

Capsules F1A, F1B, F1C, and F1D contained the bottom support piece (pedestal) with a large slot and were filled in a helium environment. Radiographic and ultrasonic examinations indicated a single sodium void in the slot below each fuel pin, plus numerous small voids ($<1/16$ inch) in the annulus between specimen and the capsule wall. * On the assumption that these voids could rise and agglomerate in the annulus, causing reduced cooling to the fuel pin, it was decided to attempt to remove these voids non-destructively using a mechanical force. **

The most effective treatment consisted of melting the sodium, centrifugation of the capsule (at Sandia Corporation), followed by slow cooling of the capsule from the bottom up. Radiography subsequently revealed that the shaking and/or centrifugation treatments had induced some fuel cracking and relocation, as well as crushing the extensometers to various degrees.

These difficulties led to revisions in the sodium filling technique and to the development of an improved procedure. By narrowing the filling slot in the pedestal at the lower end of the capsule and by filling under vacuum, the entrapment of gas bubbles was eliminated. Capsules F1E and F1F were filled in that manner. After sodium filling, the fill tubes were sealed and the end caps welded to the capsule wall. A thermal test was performed by heating the capsule to 1050°F and holding at that temperature for one hour, followed by a slow quench. The capsules were then non-destructively examined using eddy current and radiographic techniques. Post-irradiation examination of the fuel pins indicated that adequate sodium bonding was achieved in all pins.

* "Sodium-Cooled Reactors, Fast Ceramic Reactor Development Program, Twelfth Quarterly Report, July-September 1964," GEAP-4723.

** "Sodium-Cooled Reactors, Fast Ceramic Reactor Development Program, Thirteenth Quarterly Report, October 1964-January 1965," GEAP-4795.

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FIGURE B-1. CAPSULE FABRICATION FLOW SHEET

NEG. NO. 28034

FIGURE B-2. SODIUM FILLING APPARATUS

FIGURE B-3. SODIUM FILLING SYSTEM

FIGURE B-4. CAPSULE ASSEMBLY

2

APPENDIX C

GAMMA SCANNING PROFILES

APPENDIX D

PROFILOMETER MEASUREMENTS

The following information should be considered in the use of these plots.

1. Full scale on the trace represents 20 inches of travel including approximately 0.2 inch of standard.
2. The diameter of the standard is 0.242 inch.
3. The small peak observed on the bottom end of each of the fuel pins is the weld area.
4. Pre-irradiation diameter measurements were made with a micrometer and some bias may exist between a correlation of these values with the profilometer results.

FIGURE D-1. PROFILOMETER MEASUREMENTS — CAPSULE F1A

FIGURE D-2. PROFILOMETER MEASUREMENTS —
CAPSULE F1B

FIGURE D-3. PROFILOMETER MEASUREMENTS —
CAPSULE F1C

FIGURE D-4. PROFILOMETER MEASUREMENTS - CAPSULE F1D

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FIGURE D-6. PROFILOMETER MEASUREMENTS —
CAPSULE F1F

APPENDIX E

FISSION GAS ANALYSIS

TABLE E-1

GAMMA SPECTROMETRIC RESULTS

<u>Fuel Pin</u>	<u>Atoms Kr-85/cc</u>	<u>Total cc of Gas*</u>	<u>Total Atoms Kr-85</u>
F1A	1.0×10^{16}	7.6	7.6×10^{16}
F1B	5.1×10^{15}	6.9	3.5×10^{16}
F1C	5.0×10^{15}	9.6	4.8×10^{16}
F1D	7.4×10^{15}	7.3	5.4×10^{16}
F1E	1.7×10^{16}	9.4	1.6×10^{17}
F1F	2.5×10^{16}	7.8	1.9×10^{17}

* Calculated at STP.

TABLE E-2

GAS CHROMATOGRAPHIC ANALYSES
VOLUME PERCENT

<u>Fuel Pin</u>	<u>O₂</u>	<u>N₂</u>	<u>Xe</u>	<u>Kr</u>	<u>He</u>
F1A	0.1	0.45	3.25	0.5	Balance
F1B	0.08	0.23	1.6	0.22	Balance
F1C	0.07	0.31	8.7	1.4	Balance
F1D	0.1	0.5	3.1	0.35	Balance
F1E	0.65	2.05	6.35	0.8	Balance
F1F	0.2	0.5	7.3	1.33	Balance

TABLE E-3

GAS MASS SPECTROMETRIC ANALYSIS
ISOTOPIC DISTRIBUTION

<u>Isotope</u>	<u>F1E</u>	<u>F1F</u>
Kr-86	49.39 ± 0.99	47.62 ± 2.38
Kr-85	6.72 ± 0.13	6.44 ± 0.64
Kr-84	26.69 ± 0.80	27.44 ± 1.37
Kr-83	17.20 ± 0.52	18.50 ± 1.31
Xe-136	28.55 ± 1.34	26.49 ± 1.32
Xe-134	31.08 ± 1.55	33.94 ± 1.70
Xe-132	28.38 ± 2.84	24.27 ± 2.43
Xe-131	12.01 ± 1.20	15.30 ± 1.53

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