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**HIGH BURNUP PERFORMANCE
OF
POWDER AND PELLET
MIXED-OXIDE FUEL**

K. J. Perry
W. E. Baily

Approved:

J. H. Field
Principal Project Engineer

Approved:

for E. L. Zebroski, Manager
Development Engineering

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BREEDER REACTOR DEPARTMENT • GENERAL ELECTRIC COMPANY
SUNNYVALE, CALIFORNIA 94086

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ABSTRACT

The performance at high burnup (~ 148,000 MWd/Te) of powder and pellet mixed-oxide fuel was investigated. Fuel-cladding reaction resulted in the uniform loss of 0.003 to 0.004 inch of 347 stainless steel from the inside surface of the cladding at inside surface temperatures in excess of 1300°F for both types of fuel. Nonuniform cladding attack of 0.005 inch was observed in the pellet pin.

1. SUMMARY

An irradiation experiment was conducted to determine the operating characteristics of vibratory-compacted and pellet mixed-oxide fuel under burnup, linear power, and cladding temperature conditions typical of, or higher than, those expected in advanced LMFBR's. This experiment was conducted in a thermal flux and consisted of two axially aligned fuel pins of 0.250-inch diameter. The pins were clad with type 347 stainless steel and contained $(\text{Pu}_{0.25}\text{U}_{0.75})\text{O}_{2.00}$ fuel of 84% theoretical density. The pins were irradiated simultaneously at 23.5 kW/ft to a peak burnup of 149,000 MWd/Te. Cladding temperatures were in excess of 1300°F.

Post-irradiation examination showed that while both pins were intact, there was general attack on the inside surface of the cladding. Cladding reactions of up to 5 mils were observed in regions where the cladding operated at temperatures between 1300° and 1400°F. Regions of the fuel pins that operated at temperatures of 1200°F exhibited cladding attack of approximately 1 mil. Electron microprobe examinations of the reacted regions indicated the presence of Pd, Mo, Cs, and Ba.

Maximum fuel pin diametral changes of 1.2% and 0.7% were observed for the pellet and powder fuel, respectively. Center fuel melting occurred in both pins in this test and this condition is believed to be partially responsible for the observed diametral change. Best measurements of the pre-and-post-irradiation fuel volumes indicate that the fuel swelled at a rate of approximately 0.28% $\Delta V/V$ per 10^{20} f/cc. Examination of the fuel microstructures showed that thermal performance of the two fuels was equivalent at high fuel burnup. Fission gas release fractions of 91% and 78% were calculated for the pellet and powder fuel, respectively.

2. INTRODUCTION

The experimental results of a thermal flux test carried out to explore the performance of pellet and powder mixed-oxide fuels at peak cladding temperatures of 1400°F are described in this report.

The fabrication method for fast reactor fuels must be optimized to satisfy both performance requirements and low fabrication costs. Sintered pellet and vibratory-compacted powder fuel are two potential fabrication methods but their relative merits regarding applications to fast reactor technology remain uncertain. The objective of this experiment was to obtain performance data for these two fuel types under operating conditions typical of advanced LMFBR's. A second objective of this experiment was to establish the stability of type 347 stainless steel cladding in contact with mixed-oxide fuel at temperatures up to 1400°F.

The fuel pins for this test contained fuel of the same initial smear density (84% of theoretical) and the maximum exposure attained was about 149,000 MWd/te.* Information was obtained on (1) cladding deformation, (2) fuel swelling, (3) fuel redistribution, (4) fuel-cladding reaction, (5) fission-gas release, and (6) the thermal performance of both types of fuel.

3. DESCRIPTION OF EXPERIMENT

3.1 IRRADIATION CAPSULE DESIGN

The test capsule was designed to permit simultaneous irradiation of two fuel pins under similar conditions. The capsule design is shown in Figure 3-1. Two 0.250-inch diameter fuel pins were positioned one above the other in the capsule. The irradiation was performed in the General Electric Test Reactor (GETR) using a vertical and radially adjustable facility tube (V-RAFT).² The use of this facility permitted the irradiation of both pins at a constant power level.

* The results of a companion test (E5A) to determine the short-term performance of solid pellet and powder fuel have been reported previously.¹

GENERAL ELECTRIC		706375
FCR CAPSULE SERIES-E		
QTY	NAME	DESCRIPTION
1	1	NOSE PIECE 145F555 P1
1	2	TUBE 145F555 P2
1	3	BACKHEAD 886D109 P3
1	4	TUBE 886D109 P2
1	5	SPACER 886D109 P1
1	6	TUBE 1/2" O.D. WALL - 30" LG. 304 STN. STL.
2	7	END PIECE 145F555 P4
2	8	TUBE 1/2" O.D. WALL - 30" LG. 304 STN. STL.
2	9	TRANSITION 145F555 P7
2	10	END CAP 886D109 P5
2	11	BELLOWS 107C5182 G2
1	12	SPACER 886D109 P4
2	13	BELLOWS 107C5182 G1
1	14	INSULATOR 886D109 G1
1	15	SPACER 886D109 P7
2	16	DISC 1/2" O.D. WALL - 1/2" THK. 304 STN. STL.
2	17	TUBE 1/2" O.D. WALL - 2" LG. 304 STN. STL.
10	18	TUBE 1/2" O.D. WALL - 1/2" LG. 304 STN. STL.
4	19	TUBE 1/2" O.D. WALL - 1/2" LG. 304 STN. STL.
18	20	WIRE 1/2" DIA. 304 STN. STL.
1	21	FUEL PIN 107C5182 G1
1	22	FUEL PIN 107C5182 G2
1	23	WATERMOTOR CONWAY 555K-G-480 OR EQ.
2	24	T.C. BUSH 104B281 P1
1	25	SPACER 145F555 P11
1	26	LATCH 104B281 P1
1	27	BOLT 104B281 P3
1	28	ADAPTOR 145F555 P12
1	29	FLEX HOSE SEE DETAIL
1	30	PLATE 844D452 P6
1	31	UNION BUSH P3
1	32	COVER P1
2	33	BOLT P7
1	34	PLATE P5
2	35	PLATE P4
3	36	BOLT P2
2	37	BRIDGE 40-24-1/2" LG. STL.
1	38	SPACER 1/4" X 1/2" X 1/2" ALUM.
4	39	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	40	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
6	41	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
4	42	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	43	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	44	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
5	45	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	46	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	47	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	48	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	49	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM
1	50	WATERMOTOR 1/2" O.D. WALL - TYPE-B 80-100 WPM

(G) E-X-B

Figure 3-1 Design of Capsule

The pins were individually encapsulated to prevent the malfunction of one pin from affecting the operation of the other. Another design feature of the capsule was the use of bellows to accommodate the thermal expansion of the NaK thermal bond. This design significantly decreased the possibility of entrapment of gas bubbles in the NaK heat transfer medium which could result in hot spots and subsequent fuel pin failure.

Temperatures were measured radially and axially in the capsule by chromel-alumel thermocouples. These thermocouples were used to monitor the operation of the capsule and to determine the operating power.

3.2 FUEL PIN DESIGN

The fuel and cladding parameters are listed in Table 3-1. Ceramic grade powder was prepared by the co-precipitation process. High-density granules suitable for vibratory compaction were prepared by a pre-press and granulation process.³

The fuel pin design is shown in Figure 3-2. After loading, each fuel pin was backfilled with helium at one atmosphere and welded.

TABLE 3-1
FUEL PIN DESIGN PARAMETERS - CAPSULE E5B

Fuel	E5B-1	E5B-2
Type	Solid Pellet	Powder
Composition	(PU _{0.25} U _{0.75})O _{2.00}	
O/M Ratio	1.999-2.000	
% U-235/U	40	
Length, in.	4.87	4.88
Smeared Density, %	83.5 - 85.1	84.1
Pellet Density, %	85.2 - 86.8	----
Pellet Diameter, in.	0.2164-0.2183	----
Diametral Fuel-Clad Gap, in.	0.0020-0.0039	0
Insulators	UO ₂	
Density, %	95.0	82.0
Pellet Diameter, in.	0.2165	----
Fuel Cladding	Welded and Drawn	
Type	347 SS	
o.d., in.	0.251 Nom.	
Wall, in.	0.015 Nom.	
i.d., in.	0.2203±0.0001	

DRAWING NO. 107C5163

TITLE			PART NO.	
FUEL PINS				
FIRST MADE FOR F.C.R. SERIES E				
NEXT ASSEMBLY 706E374G1 & 706E375G1				
G2	G1	PT.	NAME	DESCRIPTION
1	1	1	END PLUG	SEE DETAIL
1	1	2	TRANSITION	
1	1	3	SPRING	
1	1	4	END PLUG	SEE DETAIL
1	1	5	TUBE	.717 O.D. x .656 I.D. x 1/4 LG. 347 STN. STL.
1	1	6	TUBE	250 O.D. x .220 I.D. x 6 3/4 LG. 347 STN. STL.
		7	INSULATOR	.217 DIA x 1/4 LG - UO ₂
		19	FUEL PELLETS	.217 DIA x 1/4 LG. - PuO ₂ - UO ₂
AR	AR	9	SHIM STOCK	.217 DIA x .010 THK - 304 STN. STL.
		1	10 SHIM	.217 DIA x 1/4 LG 304 STN. STL.
AR		11	INSULATOR	UO ₂ POWDER
AR		12	FUEL	PuO ₂ - UO ₂ POWDER
AR		13	PLUG	STN STL WOOL

REF.
F.C.R. FUEL PINS E-II-C1 & C2 107C5133
USE 167A1708 FOR FUEL LOADING.

2. FUEL PIN FAB. SPEC. 21-A-4954.

1. DEGREASE ALL PARTS.
NOTES:

SECTION A-A

Figure 3-2 Fuel Pin Design

Chemical analysis for the fuel and cladding are listed in Table A-1 and A-2, (Appendix A). Pre-irradiation cladding metallography is shown in Figure A-1.

3.3 IRRADIATION

The capsule was irradiated for 349 effective full power days (EFPD) at a peak power of 23.5 kW/ft and was subjected to 87 thermal cycles. Figure 3-3 illustrates the relative location of the fuel in the neutron flux. Maximum and average axial flux profiles are shown.

The capsule power history is shown in Figure 3-4. Peak operating power was approached on a ramp at the beginning of the irradiation to allow sufficient time for restructuring of the fuel.

Several methods were used to determine capsule operating power. These were (1) thermal dam thermocouples, (2) water calorimetry, (3) reactor physics calculations, and (4) fuel burnup analysis. The powers determined by these methods are listed in Table 3-2. During the irradiation the thermal dam thermocouple readings were used to control the capsule power.

TABLE 3-2
CAPSULE E5B - LINEAR POWER

As Indicated By	Peak	Average
Thermal Dam Thermocouples	20.4	19.0 kW/ft
Water Calorimetry	--	20 - 22
Reactor Physics	20.8	19.3
Fuel Burnup	23.5	22.5

Figure 3-3 Mechanical Arrangement and Irradiation Position

Figure 3-4 Linear Power History

4. POST-IRRADIATION EXAMINATION

4.1 NEUTRON RADIOGRAPHY

Neutron radiographs of the capsule were taken at the end of the irradiation. These radiographs indicated that the fuel pins were intact and that some fuel melting had occurred in both pins. In addition, in the pellet fuel pin the center void was observed to be closed in at least two locations. Subsequent metallographic examinations of these areas indicated that a fuel bridge, within the center void, did exist in two locations. Several unsuccessful attempts were made to establish the pellet interfaces by neutron radiography. The results of these radiographs indicated that at high burnups and temperatures the identification of individual pellet interfaces is lost.

4.2 GAMMA SCANS

Gamma scans were taken of the capsule prior to the destructive examination. The following energy levels were discriminated:

1. >0.670 MeV for gross scan
2. 0.513 MeV for Ru-106
3. 0.764 MeV for Zr-Nb-95
4. 1.600 MeV for Ba-La-140

A gross gamma scan of the capsule is shown in Figure 4-1.

In the pellet fuel pin gamma activity peaking at points 2D, 2E, 2F, and 2G, as shown in Figure 4-1, corresponded to the location of fuel bridges observed in the neutron radiograph.

In the powder fuel, destructive examination showed that the high gamma activity area (1F) at the bottom of the pin resulted from filling of the central void with molten fuel. The points identified on Figure 4-1 were scanned to determine the gamma intensity of specific isotopes. These results are tabulated in Appendix A, Table A-3. These data indicated that the fuel bridge regions exhibited high Ba-La-140 activity.

4.3 PHYSICAL MEASUREMENTS ON THE FUEL CLADDING

Physical measurements were performed on the fuel pins prior to their destructive examination. The appearance of the cladding was similar to unirradiated control samples and no effect from the high temperature NaK environment was observed. Decay heat kept the cladding surface at a temperature in excess of 220°F during the examination and some darkening occurred during prolonged exposure to air.

Profilometer measurements showed maximum diametral increases of 1.2% (0.0030 in.) for the pellet fuel cladding and 0.7% (0.0017 in.) for the powder fuel cladding. Axial profile measurements were made at 0-, 45- and 90-degree orientations and a helical trace was obtained with the specimens rotating. Profilometer traces are shown in Figures 4-2 and 4-3.

Diametral increase was nonuniform over the length of the pellet fuel cladding. The helical trace indicated a maximum ovality of 0.006 inch. The two areas of maximum ovality corresponded to the axial location of maximum strain on the 45- and 90-degree traces. Diameter reductions of up to 0.001 inch were observed on the 0-degree trace. The large ovality in the pellet fuel cladding indicates that most of the diameter increase resulted from cladding distortion rather than diametral strain. The maximum ovality measured on the powder fuel cladding was 0.002 inch. No reduction in diameter similar to that observed on the 0-degree trace of the pellet fuel pin was found.

Fuel pin diametral expansion showed no correlation with fuel-cladding gap or fuel density. Axial plots of these parameters for the pellet pin are shown in Appendix A, Figure A-2.

Length measurements on both fuel pins indicated that no significant change in fuel pin length occurred during irradiation.

4.4 FISSION GAS ANALYSIS

Following collection of the helium-fission gas mixture it was determined that the pellet fuel released 99 cc of fission gas and the powder fuel released 84 cc. Fractional releases of 91% (pellets) and 78% (powder) were calculated using the gas chromatography data*. Aliquots of the fission gas were analyzed by gas chromatography, gamma counting, and mass spectrometer. The results of these analyses are shown in Appendix A, Table A-4. Fission gas calculations are contained in Appendix B-1.

4.5 FUEL BURNUP

Burnup analysis was performed by mass spectrometry on two samples from each fuel column. Both the heavy element ratio and neodymium 148 to heavy element ratio techniques were used.⁴ The results of these analyses indicated that the peak power in the fuel was 15% higher than had been estimated by GETR physics and heat transfer

* A second calculation utilizing Kr-85 gamma counting yielded values of 100% (pellets) and 122% (powder). These values were considered in error because of the higher than theoretical release rate.

Figure 4-1a Gross Gamma Scan

calculations. Isotopic contents of the fuel before and after irradiation are given in Appendix A, Table A-5, with the results of the burnup analysis. Burnups based on neodymium 148 to heavy element ratio for the high power ends of the fuel columns differ by 21,000 MWd/Te (159,000 versus 138,000). Subsequent post-irradiation examination of the specimen utilized for burnup analysis in the peak power region of the pellet pin indicated that this section (section H 159,000 MWd/Te) contained a large central void while the powder fuel specimen (section B, 138,000 MWd/Te) did not. Flux depression calculations showed that the difference in linear power in these highly enriched fuels would be less than 1% when both were exposed to the same thermal neutron flux. Because of the flux depression, the fuel in the center of the powder specimen was of low burnup which tended to depress the average burnup value. The pellet specimen contained no fuel in the central area and this tended to enhance the burnup value. Therefore the discrepancy in the reported burnups is believed to have resulted from a physical difference in the fuel sample geometry rather than from the fuels operating at different power levels. After considering these factors it was decided that the average of the two burnups was the more realistic value (149,000 MWd/Te). On this basis, the peak power was calculated to be 23.5 kW/ft.

4.6 FUEL SWELLING

Fuel column length increases of 8% (pellet) and 3% (powder) were measured from neutron radiographs and were confirmed during the destructive examination. These values, along with the post-irradiation diameter measurements, were utilized in estimating fuel swelling. These calculations showed that the fuel occupied less volume* after irradiation than before for both the powder and pellet pin. On this basis the pellet fuel occupied 96.4% of the pre-irradiated volume and the powder fuel occupied 95.8%. This result indicates that fuel densification exceeded fission product swelling by 3% to 4%. This is reasonable since the initial fuel density was only 84% and the linear power of this experiment was high enough to cause gross sintering *in situ*.

* Fuel volume used in these calculations included internal fuel porosity minus the volume of the central void.

Figure 4.1b Gross Gamma Scan

Figure 4-2 Pellet Fuel Pin Profilometry

Figure 4-3 Powder Fuel Pin Profilometry

5. DESTRUCTIVE EXAMINATION

5.1 FUEL SECTIONING

The fuel pins were sectioned into specimens 1/2 inch to 5/8 inch in length. Macrographs were taken at both ends of each section and typical macrographs are shown with neutron radiographs in Figures 5-1 and 5-2.

Fuel bridges are visible in the central void of the pellet fuel in sections C, E, F, and G, Figure 5-1. The central void volume was only 3% at the bottom (low power) end of the pin and increased to 28% at the upper end of the pin.

Near the bottom (high power) end of the powder fuel no central void existed. Subsequent metallographic examination revealed a molten core in this region. Above this molten core two small spheres of fuel were found in the central void (see macrograph C, Figure 5-2). These spheres had a glassy appearance and were firmly attached to the wall of the void.

5.2 METALLOGRAPHIC EXAMINATION

Figures 5-3 and 5-4 show comparable sections from near the axial center of the powder and pellet fuel (E sections of Figures 5-1 and 5-2). No fuel melting occurred in these areas and these sections are estimated to have operated under equivalent conditions of power and burnup. For this reason they were used in making performance comparisons of the powder and pellet fuel. A description of these specimens is given below.

5.2.1 Description of Comparable Sections of Powder and Pellet Fuels

Irradiation Conditions:

Linear Power	22.5 kW/ft
Fuel Center Temperature	2600°C
Fuel Surface Temperature	1230°C
Cladding Inside Surface Temperature	721°C (1330°F)
Central Void Volume	Powder 9.5%; Pellets 10.3%

The central void in powder fuel section E (Figure 5-2) occupied 9.5% of the fuel area with the columnar grains extending to 80% of the fuel radius. Equiaxed grains of decreasing size extended to 90% of the fuel radius. The fuel between the equiaxed grains and the cladding was extremely porous. Pore sizes up to 0.0015 inch in diameter were measured (see Figure 5-5). Fission gas probably formed the majority of these pores but some may have contained metallic fission products which were lost during metallographic preparation. In contrast, the porosity and metallic fission products contained in the region of equiaxed grain growth were generally less regular in form and appeared to follow the grain boundaries. Metallic fission products were dispersed throughout the fuel but the greatest concentration was observed in the area of equiaxed grain growth. Stringers of white metallic fission products were visible within radial cracks. Some of these extended from the cladding through the equiaxed grain region. Metallic fission products in the region of columnar grain growth were spherical, probably as a result of surface tension effects since they were molten during irradiation.

A band of second phase material about 0.001 inch thick was located between the fuel and cladding, Figure 5-5. Although a distinct boundary existed between this material and the cladding, it is believed that this band resulted from cladding reaction rather than fission-product deposition. The inner 0.003 to 0.004 inch of the cladding showed an affected zone with distinguished grain boundaries. A light oxalic acid electrolytic etch was sufficient to remove both the band of material between the fuel and cladding and also the material within the grain boundaries of the affected zone in the cladding (see Figure 5-5). Additional etching produced the cladding structure shown in Figure 5-6*. Metallic fission products in the adjacent fuel were not affected by the etch. Intergranular precipitation of carbides in the cladding was observed in all cross sections. Regions adjacent to the affected zone exhibited both intergranular and intragranular precipitates.

The fuel morphology in pellet fuel section E, shown in Figure 5-4, was generally similar to that of the equivalent powder fuel specimen previously described. The central void occupied 10.3% of the fuel area, with columnar grains extending to 83% and equiaxed grains of decreasing size extending to 93% of the fuel radius. Comparable micrographs of the pellet and powder fuel adjacent to the cladding are shown in Figure 5-7.

* The voids in the etched cladding sample (Figure 5-6) were subsequently determined to represent locations of sigma phase removed by the oxalic acid etch. Specimens etched with Vilellas reagent confirmed this observation (see Figure 5-9).

Figure 5-1 Pellet Fuel Sections and Neutron Radiograph

Figure 5-2 Powder Fuel Sections and Neutron Radiograph

POWDER FUEL, SECTION E

30 X

LINEAR POWER	22.5 kW/ft
FUEL CENTER TEMPERATURE	2600°C
FUEL SURFACE TEMPERATURE	1230°C
CLAD INSIDE SURFACE TEMPERATURE	720°C (1330°F)
VOID VOLUME	9.5%

Figure 5-3 Structure of Powder Fuel, Powder Fuel Section E

PELLET FUEL, SECTION E

30X

LINEAR POWER	22.5 kW/ft
FUEL CENTER TEMPERATURE	2600°C
FUEL SURFACE TEMPERATURE	1220°C
CLAD INSIDE SURFACE TEMPERATURE	720 °C (1330°F)
VOID VOLUME	10.3%

Figure 5-4 Structure of Pellet Fuel, Pellet Fuel Section E

POLISHED

500X

0.001 inch

LIGHT ETCH
10% OXALIC ELECTROLYTIC

500X

Figure 5-5 Fuel-Cladding Interface, Powder Fuel Section E

POLISHED

250X

0.001 inch

ETCHED
10% OXALIC ELECTROLYTIC

250X

Figure 5-6 Polished and Etched Cladding, Powder Fuel Section E

POLISHED

POWDER FUEL

100X

0.005 inch

POLISHED

PELLET FUEL

100X

Figure 5-7 Comparison of Powder and Pellet Fuel-Cladding Reaction at Equivalent Cladding Temperatures

The prominent feature of the pellet fuel specimen was the fuel-cladding reaction zone. This consisted of a uniform ring of metallic-appearing material, generally 0.003 to 0.004 inch thick but with local thickness of up to 0.006 inch. One specimen was etched with Vilellas reagent to detect sigma phase in the type 347 stainless steel cladding. Figure 5-8 shows micrographs which illustrate the effect of this etchant on the cladding and reaction zone. Figure 5.9 shows a higher magnification of the cladding etched for sigma phase.

Autoradiographs of the powder and pellet fuel specimens (Figures 5-3 and 5-6) may be found in Appendix C-7. Enhanced β - γ activity and decreased α activity were noted adjacent to the cladding where very high fuel burnup occurred.

5.2.2 Electron Microprobe Examination

An electron microprobe examination was performed on the opposite face (Figure 5-10) of the pellet fuel section described in subsection 5.2.1. Details regarding the instrument and experimental procedure used in the examination may be found in Reference 5.

A qualitative determination of the distribution of cladding elements and fission products in the fuel-cladding reaction zone was made. Figure 5-11 is a 500X micrograph of the area examined showing the 0.004 to 0.005 inch thick reaction zone. A specimen current image display is also shown for orienting particular features in the $K\alpha$ x-ray pulse image displays shown in subsequent figures. Comparisons may be made by utilizing the grid shown on the displays. Key grid points are marked on the photographs. It is noted that the specimen current and x-ray image displays are right to left mirror images of the micrograph and are 425X magnification. $K\alpha$ x-ray pulse images for the primary cladding constituents Fe, Ni, and Cr are shown in Figure 5-12. Stratification of these elements is evident. A quantitative determination of the relative Fe, Ni, and Cr concentrations was made at points across the reaction zone. The composition at each point going from the fuel side to the cladding side is shown in Figure 5-13.

Iron appeared to be the most mobile of the cladding constituents and was found combined with metallic fission products as far into the fuel as the equiaxed grain region. Free iron was found in the fuel matrix in proximity to the reaction zone. Cr was detected combined with fission products in the fuel near the fuel-cladding reaction zone. Pd, Mo, Ba, and Cs were also detected in the fuel-cladding reaction zone. The highest concentrations of these fission products were found adjacent to the cladding base metal. Figures 5-14 and 5-15 qualitatively show the concentrations of these elements. A specimen current image is included because the location of the grid is slightly offset from that of Figure 5-11.

Small islands of fuel were found in the reaction zone. These appear as grey inclusions in the micrograph and as black spots on the specimen current image, Figure 5-11.

Analysis of two metallic agglomerations in the outer fuel region (outside of the columnar grains) indicated the presence of Mo, Ru, and Tc in the ratio of 1.0, 0.4, and 0.25. Metallic fission products in the columnar grain region contained Mo and Ru in approximately equal quantities along with traces of Tc and Rh. Regions in the fuel matrix adjacent to the metallic fission products were analyzed. The only fission product detected was a trace of Zr.

A grey phase in the equiaxed grain region of the fuel matrix was examined and Zr, Ba, Cs, and Sr were found. It was determined that the spacial distribution of Ba and Zr were identical and coincided with the second phase region. The Cs rich areas were in proximity with, but not identical to, the Ba-Zr rich areas. The Sr was barely detectable and the exact distribution could not be determined. Although this grey phase had an appearance similar to the alumina found in a control sample, no Al was detected. It is plausible that the second phase is barium zirconate.

5.2.3 Other Metallographic Specimens

Sections A and H of the pellet fuel and sections A and B of the powder fuel are described in Appendix C. Micrographs and autoradiographs of these specimens are presented there as well as autoradiographs of the E sections previously described. These results are summarized below.

Pellet fuel section A (Figure C-1) from the low power end of the pellet fuel had a calculated cladding inside surface temperature of 1220°F or 110°F lower than the E sections previously described (subsection 5.2.2). It is significant that except for an isolated penetration of 0.002 inch, very little fuel-cladding reaction occurred at this cladding temperature.

Pellet fuel section H, (Figure C-2) came from the high power end (top end) of the fuel and had a cladding inside surface temperature of 1400°F. A cladding thickness reduction of 0.004 inch occurred. Molten fuel from this section was relocated into section A above.

Figure 5-8 Pellet Fuel Section E Micrographs

VILELLAS ETCH
TO SHOW SIGMA PHASE

CLADDING OUTSIDE SURFACE TEMPERATURE = 1200°F
CLADDING INSIDE SURFACE TEMPERATURE = 1330°F

—|—
0.001 inch

ELECTROLYTIC ETCH
TO SHOW SIGMA VOIDS

500X

Figure 5-9 Sigma Phase in 347SS Fuel Cladding, Pellet Fuel Section E

SECTION D

Figure 5-10 Electron Microprobe Specimen, Pellet Fuel Section D

500X

MICROGRAPH

AREA A

MIRROR IMAGE

0.001 inch

425X

SPECIMEN CURRENT IMAGE

Figure 5-11 Area of Fuel-Cladding Reaction Zone Examined with Electron Microprobe

Fe K α X-RAY PULSE IMAGE

Cr K α X-RAY PULSE IMAGE

0.001 inch

Ni K α X-RAY PULSE IMAGE

Figure 5-12 Locations of Fe, Ni and Cr in the Fuel-Cladding Reaction Zone

Fe, Cr, AND Ni CONCENTRATION AT SELECTED
POINTS IN THE CLADDING-FUEL REACTION ZONE

<u>POINT</u>	<u>% Fe</u>	<u>% Cr</u>	<u>% Ni</u>	<u>TOTAL</u>
1	58.5	15.5	5.3	75.3
2	30.8	11.0	2.0	43.8
7	30.5	9.0	2.4	41.9
3	37.5	12.0	2.8	52.3
4	52.3	3.0	4.6	59.9
5	37.3	19.0	3.0	59.3
6	37.0	17.9	2.6	57.6
CLAD MATRIX	67.7	17.4	8.9	94.0

0.001 inch

SPECIMEN CURRENT IMAGE (LEFT PHOTOGRAPH) SHOWING THE BEAM POSITIONS (RIGHT PHOTOGRAPH) USED IN POINT COUNTING FOR Fe, Ni, AND Cr. THE IMAGES CAN BE MATCHED BY USE OF THE SUPERIMPOSED GRID MARKINGS. THE FUEL IS AT THE TOP OF THE IMAGE.

Figure 5-13 Fe, Ni, and Cr at Selected Points in the Fuel-Cladding Reaction Zone

SLIGHTLY DIFFERENT POSITION

SPECIMEN CURRENT IMAGE

Pd $L\alpha$ RATE OUT X-RAY IMAGE

Figure 5-14 Distribution of Pd in the Fuel-Cladding Reaction Zone

Mo $L\alpha$ X-RAY RATE OUT IMAGE

Cs $L\alpha$ X-RAY RATE OUT IMAGE

0.001 inch

Ba $L\alpha$ X-RAY RATE OUT IMAGE

Figure 5-15 Distribution of Mo, Cs, and Ba in the Fuel-Cladding Reaction Zone

Powder fuel section B (Figures C-3 and C-4) came from the high-power (bottom) end of the powder fuel and exhibited a classical molten structure which was used to make $\int kdt$ to melt calculations. The power and temperatures were the same as pellet fuel section H (above) and again significant fuel-cladding reaction occurred.

Powder fuel specimen A (Figure C-5) is a longitudinal section the top of which was adjacent to powder fuel section B, above. Although extensive fuel-cladding reaction occurred at the upper end of this specimen where the cladding inside surface temperature was about 1400°F, a sharp reduction in cladding attack occurred as axial heat transfer apparently reduced the cladding temperature toward the bottom of the pin. The sharp reduction in cladding attack suggests that a threshold temperature for this reaction exists. It was not possible to determine the temperature at which this occurred from this specimen. However, little reaction occurred at a cladding inside surface temperature of 1220°F in pellet fuel specimen A, while reaction occurred in all areas where cladding inside surface temperatures were in excess of 1300°F. It is probable therefore, that a temperature near 1300°F is required for the reaction to proceed at a significant rate.

Both alpha and β - γ autoradiographs of the fuel sections are shown in Figures C-6 through C-9. A brief discussion is included in Appendix C-6.

6. DISCUSSION OF RESULTS

Microstructural similarities and $\int kdt$ calculations lead to the conclusion that the pellet and powder fuels exhibited similar thermal performance. Sections from the pellet and powder fuel which were irradiated at the same power had similar columnar radii. These were 82% of the fuel radius in the pellet fuel and 80% of the radius in the powder fuel. Central voids of 32% existed in both fuels. In the molten fuel sections the powder fuel showed a molten radius of 46%. A central void of 54% existed in the comparable pellet specimen where molten fuel had slumped downward. The columnar grain radii in these two sections were similar, 81% for the pellets and 79% for the powder.

The powder fuel specimen with its well defined molten and columnar grain radii provided data for quantitative calculation of the thermal characteristics for high burnup mixed oxide fuel. The calculational technique of Robertson⁶ was used to calculate surface-to-melting,

$$\int_{T_s}^{T_m} kdt$$

and surface-to-columnar grain,

$$\int_{T_s}^{T_{cg}} kdt$$

integrated thermal conductivities. The values calculated for each integral were:

$$\int_{T_s}^{T_m} kdt = 33 \text{ watts/cm}$$

$$\int_{T_s}^{T_{cg}} kdt = 19.6 \text{ watts/cm}$$

Where:

- T_m = melting temperatures
- T_{cg} = temperature of columnar grain growth
- T_s = fuel surface temperature

TABLE 6-1

THERMAL IRRADIATION PROPERTIES FOR PELLET AND POWDER FUEL CAPSULE E5B

Fuel Section	Linear Power (kW/ft)	Inside Surface Temperature, (°F)	Cladding Fuel Surface Temperature, T_s (°C)	Thermal Conductance Btu (h-ft ² -°F)	$\int_{T_s}^{T_c} \frac{k dt}{T_s}$ (W/cm)	Fuel Center Temperature, T_c (°C)
B Pellet	20.4	1220	1360	960	27.7	2640
E Pellet	22.5	1330	1230	1470	30.0	2580
F Pellet	22.9	1360	1350	1235	28.1	2610
G Pellet	23.5	1390	1280	1535	25.2	2420
B Powder	23.5	1390	1250	1630	33.0	2750 (Molten)
E Powder	22.5	1330	1230	1470	30.0	2580

A fuel melting temperature of 2750°C* was assumed to calculate a columnar grain growth temperature of 2150°C and a fuel surface temperature of 1250°C. Utilizing these values a fuel-to-cladding gap conductance of 1630 Btu/h-ft² °F was calculated using the $\int k dt$ curve of Craig and Baily.⁸ The calculated columnar grain growth temperature of 2150°C agrees closely with values reported by Hausner and Nelson⁹ and by Potter and Elyard¹⁰ for UO₂. The resulting value of 1630 Btu/h-ft² °F for gap conductance is consistent with the results of Craig et al.¹¹

Because no molten radius existed in all powder or pellet fuel samples, calculations were based on the columnar grain growth radius assuming a temperature of 2150°C for columnar grains to initiate. The results of these calculations are tabulated in Table 6-1.

With the exception of one low value the gap conductances calculated for this experiment range from 1235 to 1630 for linear powers of 22.5 to 23.5 kW/ft. Four of these five values fall in the narrow range of 1470 to 1630 Btu/h-ft² °F. These values of gap conductance include any effect which results from the presence of the fuel-cladding reaction zone. The results from these experiments indicate that the effective gap conductance between the fuel and cladding including the reaction zone is comparable to pins where no reaction zone is present.

The high fission gas release fractions observed in this experiment are comparable to other gas release data from high burnup mixed oxide fuels irradiated with fuel center temperatures in the range of 2300° to 2600°C. Typical gas release results from other thermal flux experiments are noted in Table 6-2.

Fuel volume measurements[†] after irradiation showed that the fuel volume decreased by 3% to 4% during irradiation. Post-irradiation microstructures confirmed that gross densification of the fuel occurred during the irradiation, resulting in a lower fuel volume after the test. These results indicate that the 84% smeared density fuel used in these experiments was sufficiently low to result in minimal mechanical fuel-cladding interaction. Fuel swelling rates calculated from these pins indicate a rate of 7% per 100,000 MWd/Te or 0.28% $\Delta V/V$ per 10²⁰ f/cc, assuming a post-irradiation fuel density of 97% theoretical.

Cladding inside surface temperatures in the 1330°-1400°F range resulted in general fuel-cladding reaction (or fission product-cladding reaction) to a depth 0.003 to 0.004 inch with both powder and pellet fuel. A summary of the depth of cladding reaction is shown in Table 6.3. Although the deepest isolated penetration (0.0051 inch) was found on a pellet fuel specimen, the general appearance of comparable sections indicates equivalent depths of generalized attack in powder and pellet fuel cladding. Temperature dependence of the cladding reaction was confirmed by the finding of only localized reaction to a depth of 0.002 inch in a specimen with cladding inside surface temperature of 1220°F.

* Out-of-pile melting point experiments by Krankota and Craig⁷ indicate that the melting point of mixed-oxide fuel is depressed by 50° to 100°C at burnups in excess of 100,000 MWd/Te.

† The volume determinations were made from fuel-blanket interface micrographs and cross-section cuts through the fuel pin and included axial fuel growth and center void volumes.¹²

Out-of-pile compatibility experiments by T. Lauritzen at 1400°F for 1000 hours resulted in no fuel-cladding reaction for stoichiometric and hypostoichiometric fuel.¹⁸ Only slight attack from hyperstoichiometric fuel was found in these 1000-hour tests. It is probable, therefore, that the presence of fission products contributed to the substantial reaction observed in the present work. The mechanism may be through grain boundary attack since whole grains were observed adjacent to, but disconnected from, the cladding base metal.

Mechanisms associated with this cladding reaction suggest that oxygen may play a role in these reactions. For example, oxygen is liberated when the metals in mixed-oxide fuels are consumed by fission. Much of this oxygen will combine with the fission fragments, depending upon which species is fissioned, the flux spectrum, the temperature and stoichiometry of the fuel. Some of the liberated oxygen will be available to raise the O/M ratio.¹⁹ Thermodynamic analysis indicates that when the O/M ratio reaches 2.0, (or perhaps even a slightly lower value) the chromium in stainless steel should begin to oxidize. Thus, the fuel should become more aggressive as burnup proceeds and the oxygen potential in the fuel increases. It is well known, however, that stainless steel forms a protective film which effectively prevents oxidation in many environments where extensive reaction would be thermodynamically predicted. This is supported by the results of out-of-pile tests where very little reaction is observed, even with hyperstoichiometric fuels.

Low melting temperature fission-product metals may also play a part by liquid metal attack. Antimony, for example, is particularly aggressive in this respect. Tellurium, for which antimony is a precursor, has often been found in the fuel-cladding gap.

As additional information is obtained from high cladding temperature tests and related out-of-pile experiments, it is reasonable to expect that a clear understanding of the mechanisms associated with cladding-fuel reactions will be developed. Results from these efforts should provide the necessary information for the commitment of near term LMFBR's. Advanced LMFBR's that will be operated at higher power levels and temperatures will require continuing development work to establish means for minimizing or eliminating these reactions.

TABLE 6-2

FISSION GAS RELEASE DATA FROM THERMAL FLUX IRRADIATIONS

Fuel Pin	O/M Ratio	Smeared Density	kW/ft	Burnup MWd/Te	% Fission Gas Release
E1E ⁽¹³⁾	2.00	91	20	87,000	84
E1F ⁽¹³⁾	2.00	92	17/31	75,000	96
E2C-1 ⁽¹⁴⁾	2.00	82	20	117,000	89
E2C-2 ⁽¹⁴⁾	2.00	91	20	108,000	97
E2D-1 ⁽¹⁵⁾	2.00	87.5	16	136,000	80
E2D-2 ⁽¹⁵⁾	1.96	88.3	16	133,000	81
E2F ⁽¹⁶⁾	2.00	95.4	-	242,000	83
E2G ⁽¹⁶⁾	2.00	96.5	-	208,000	82
E2H ⁽¹⁶⁾	2.00	93.5	-	322,000	87
E2J-1 ⁽¹⁷⁾	1.98	89	20	124,000	73
E2J-2 ⁽¹⁷⁾	1.98	89	20	125,000	79
E5B-1	2.00	84	23	148,000	91
E5B-2	2.00	84	23	148,000	78

TABLE 6-3

FUEL CLADDING REACTION MEASUREMENTS

Specimen ^a Number	Cladding Wall Thickness (in.)	Reduction in Cladding Wall Thickness (in.)	Cladding Wall Plus Reaction Layer (in.)	Reaction Layer Thickness (in.)	Calculated Cladding Inside Surface Temperature (°F)
Control	0.0153 ^b	0	--	0	--
	0.0152 ^b	0	--	0	--
	0.0151 ^b	0	--	0	--
Powder					
A	0.0120	0.0032	0.0170	0.00050	1400
E	0.0118	0.0034	0.0161	0.0043	1330
	0.0128	0.0024	0.0161	0.0038	
	0.0136	0.0016	0.0166	0.0030	
	0.0140	0.0012	0.0165	0.0025	
Pellet					
B	0.0132	0.002	0.0157	0.0025	1220
	0.0152	0	--	0	
E	0.0112	0.0040	0.0196	0.0084	1330
	0.0129	0.0023	0.0178	0.0049	
	0.0136	0.0016	0.0176	0.0040	
	0.0136	0.0016	0.0184	0.0048	
F	0.0101	0.0051	0.0164	0.0063	1360
	0.0111	0.0050	0.0164	0.0053	
	0.0120	0.0032	0.0178	0.0058	
	0.0121	0.0033	0.0168	0.0047	
	0.0125	0.0027	0.0169	0.0044	
	0.0136	0.0016	0.0172	0.0036	
G	0.0116	0.0036	0.0178	0.0062	1390
	0.0125	0.0027	0.0169	0.0044	
	0.0139	0.0013	0.0164	0.0025	

a Refers to Figures 5.1 and 5.2

b Control measurements on unaffected cladding agree with pre-irradiation measurements.

7. CONCLUSIONS

The results of this experiment provided the first correlation of fuel-cladding reaction with temperature under this program. More specifically, it was observed that type 347 stainless steel cladding operating at temperatures in excess of 1300°F reacted with irradiated mixed-oxide fuel to a uniform depth of 3 to 4 mils of the cladding wall thickness in both pellet and powder pins. Localized cladding reaction of up to 5 mils was observed in the pellet pin.

A comparison of these observations on cladding reaction with other mixed-oxide fuel tests is difficult because of the high cladding temperatures that these pins experienced. The amount of reaction observed in the low temperature regions of these pins (~1220°F cladding temperature) is 2 mils. This value is consistent with other observations on types 316, 304, and I-800 where the spread in reaction depth ranged from 0.5 mil to 3 mils.²⁰ No comparable data are available from other experiments to relate to the high temperature results.

Fuel volume changes during irradiation indicated that both the powder and pellet fuel at comparable smeared densities exhibited a growth rate of approximately $0.28\% \Delta V/V$ per 10^{20} f/cc assuming a post-irradiation fuel density of 97% theoretical. Fuel pin diameter changes that occurred during irradiation were 1.2% and 0.7% for the pellet and powder pin, respectively. The thermal performances of the two fuel types, as evidenced by the post-irradiation microstructure, indicated comparable end-of-life thermal conductivity and gap conductance. Gap conductance values of 1470 to 1630 Btu/h-ft² °F were calculated.

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 W. V. Cummings - Microprobe examination
 R. E. Smith - Metallography

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APPENDIX A
FABRICATION AND SELECT
POST IRRADIATION DATA

TABLE A-1
FUEL IMPURITY ANALYSIS - CAPSULE E5B

	Pellets (ppm)	Powder (ppm)
Ag	1	--
Al	5	20
As	--	--
B	1	--
Be	--	--
Bi	--	--
Ca	10	100
Cd	--	--
Cr	10	100
Cu	1	2
Fe	194	490
Ge	--	--
K	10	--
Li	--	--
Mg	20	20
Mo	2	10
Mn	10	20
Na	20	20
Ni	10	50
P	--	--
Po	1	--
Si	100	200
Sn	1	5
Ti	200	200
Tl	--	--
V	--	--
Zn	--	--
Total Metallic Impurity	596	1237
C	95	100
Cl	8	6
F	<1	8
N	100	34

TABLE A-2

**CLADDING CHEMICAL ANALYSIS AND MECHANICAL
PROPERTIES DATA**

Impurity Analysis		Room Temperature Tensile Test	
Element	Percent	347 SS Cladding Welded and Drawn	
C	0.05	Tensile Strength (psi)	105,000
Mn	1.16	Yield Strength (psi)	51,000
P	0.023	Percent Elongation	39
S	0.009	Grain Size (ASTM)	6-8
Si	0.72		
Ni	9.32		
Cr	18.20		
Cb-Ta	0.91		
Mo	0.14		
Cu	0.20		
B	0.0012		
N ₂	0.056		

TABLE A-3

RELATIVE ACTIVITY OF ISOTOPES - CAPSULE E5B

Position ^a Number	ZrNb-95 (Relative) ± 15%	La-140 (Relative) ± 15%	Other Isotopes Observed
1A'	150		Co-60
1A	175		Co-60, Cr-51
1B	2,600	180	Cs-137, Ru-103
1C	11,000	1,700	Ru-103
1D	7,500	1,200	Ru-103 (High)
1E	7,000	1,000	Ru-103
1F	11,000	1,700	Ru-103
2A'	475		
2A	350		Co-60, Cr-51
2B	<1,100	80	I-131, Cs-134, 137, Cs-136
2C	7,000	450	Ru-103 (High)
2D	27,000	4,800	Ru-103
2E	15,000	3,300	Ru-103
2F	11,000	2,500	Ru-103
2G	10,000	2,300	Ru-103
2H	7,000	900	Ru-103
2I	10,000	2,100	Ru-103

^a Refers to Figure 4-1

TABLE A-4

FISSION GAS ANALYSIS - CAPSULE E5B

	Pellets	Powder
A. Total Gas Volume:	104 cc	93 cc
B. Fission Gas Volume:	99 cc	86 cc
C. Fission Gas Release:	91%	78%
D. Gas Chromatograph:	%	%
Ar + O ₂	Trace	0.4
N ₂	0.10	0.5
CO	0.04	Not Detected
CH ₄	Not Detected	Not Detected
CO ₂	Not Analyzed	Not Analyzed
Xe	85.5	81.7
Kr	8.71	8.55
He	4.85	7.40
H ₂	0.18	Not Detected
E. Gamma Counting		
Total Atoms Kr-85	1.6×10^{19}	1.9×10^{19}
F. Mass Spectrograph:	%	%
Kr-83	12.26	13.16
84	30.04	30.16
85	6.59	6.86
86	51.11	49.69
Xe-131	10.60	11.12
132	18.82	18.51
134	28.17	28.35
136	42.41	42.01
Xe	92.1±3%	89.9±3%
Xe + Kr		

TABLE A-5

BURNUP AND ISOTOPIC DISTRIBUTION - CAPSULE E5B

		Pellets		Powder	
A. Burnup from Neodymium 148/ Heavy element ratio MWd/Te		High Power	Lower Power	High Power	Lower Power
		159,000 ±6,000	130,000 ±4,900	138,000 ±5,200	115,000 ±4,300
B. Burnup from Heavy Isotope Ratios					
MWd/Te		145,000 ±18,000	122,000 ±15,800	138,000 ±17,900	110,000 ±14,300
C. Post-Irradiation Isotopic Content (Atom %)					
U-234		0.47	0.49	0.42	0.48
U-235		30.40	31.84	29.82	32.16
U-236		2.29	1.88	2.00	1.61
U-238		66.84	65.78	67.76	65.76
Pu-239		67.95	72.45	70.52	74.46
Pu-240		27.34	23.84	25.34	22.25
Pu-241		4.02	3.25	3.56	2.90
Pu-242		0.68	0.47	0.57	0.39
Pu-239		24.70	28.40	24.00	26.90
U-238					
D. Pre-Irradiation Isotopic Content (Atom %)					
U-234			0.47		0.47
U-236			0.11		0.11
U-235			40.10		38.10
U-238			59.90		61.90
Pu-239			90.91		91.00
Pu-240			8.36		8.30
Pu-241			0.69		0.70
Pu-242			0.03		0.04
Pu-239			49.40		47.70
U-238					
Pu			24.50		24.50
U

HCl-H₂O₂ ETCH

250X

Figure A-1 Metallography of Type 347 Stainless Steel Cladding

Figure A-2 Axial Plot of Parameters Which May Affect Cladding Diametral Expansion

APPENDIX B

FISSION GAS CALCULATIONS

1. TOTAL FISSION GAS + ARGON FILL GAS COLLECTED:

<u>Pellets</u>	<u>Powder</u>
104 cc	93.3 cc

2. CALCULATION OF TOTAL QUANTITY OF FISSION GAS GENERATED:

- A. Fuel burnup based on Nd-148 - heavy metal ratio analysis, MWd/Te
Average of four analyses = 136,000 MWd/Te

- B. Mass of heavy metal per pin

	<u>Pellets</u>	<u>Powder</u>
gms oxide	28.16	28.30
gms metal	24.81	24.93

- C. For pellets (200 MeV/fission)

$$136,000 \text{ MWd/Te} \times \frac{2.7 \times 10^{15} \frac{\text{fission}}{\text{gm (U+Pu)}}}{1 \text{ MWd/Te}} \times 24.81 \text{ gm (U+Pu)}$$

$$\frac{0.32 \text{ atom gas}}{\text{fission}} \times \frac{22,400 \text{ cc}}{6.023 \times 10^{23} \text{ atoms}} = 108 \text{ cc fission gas generated}$$

For Powder = 108 cc

3. CALCULATION OF FISSION GAS % RELEASED:

A. From gas chromatography

	<u>Pellets</u>	<u>Powder</u>
Kr %	8.71	8.55
Xe %	85.5	81.7

Calculation of fission gas quantity collected:

For Pellets

$$(104) (0.1087 + 0.855) = 98.0 \text{ cc released}$$

For Powder

$$(93.3) (0.086 + 0.817) = 84.3 \text{ cc released}$$

Calculation of % gas released:

For Pellets

$$\frac{98.0}{108} = 91\%$$

For Powder

$$\frac{84.3}{108} = 78\%$$

B. From Gamma Spectroscopy:

Total Kr-85/cc of gas collected, atoms/cc

For Pellets

$$1.54 \times 10^{17}$$

For Powder

$$2.10 \times 10^{17}$$

1. Calculation of total atoms Kr-85 collected:

For Pellets

$$1.54 \times 10^{17} \frac{\text{atoms}}{\text{cc}} \times 93.3 \text{ cc} = 1.60 \times 10^{19} \text{ atoms Kr-85}$$

For Powder

$$2.10 \times 10^{17} \frac{\text{atoms}}{\text{cc}} \times 93.3 \text{ cc} = 1.96 \times 10^{19} \text{ atoms Kr-85}$$

2. Calculation of Kr-85 yield, %

From fuel analysis:

$$\frac{\text{Pu}}{\text{U} + \text{Pu}} = 0.245$$

$$\frac{\text{Pu-239}}{\text{Pu}} = 0.909$$

$$\frac{U}{U + Pu} = 0.755$$

$$\frac{U-235}{U} = 0.401$$

$$\text{Fraction U-235} = (0.755) (0.401)$$

$$= 0.302$$

$$\text{Fraction Pu-239} = (0.245) (0.909) = 0.223$$

Relative probability of fission of U-235 vs Pu-239

$$= (0.302) (572 \text{ Barns}) = 172$$

$$\text{For Pu-239 } (0.223) (960) = 214$$

Relative fission yields of Kr-85 (per Katcoff, 1964)

$$Pu-239 = 0.099\%$$

$$U-235 = 0.273\%$$

Average probability of Kr-85 yield of Pu-239 vs U-235

$$0.099 \times \frac{214}{172 + 214} = 0.055\%$$

$$0.273 \times \frac{172}{172 + 214} = 0.122\%$$

TOTAL	0.177%
-------	--------

$$\text{Kr-85 Yield} = 0.177\%$$

$$= 0.00177 \text{ atom/fission}$$

For Pellets

$$1.60 \times 10^{19} \text{ atoms Kr-85} \times \frac{0.32 \text{ atom gas}}{0.00177 \text{ atom Kr-85}} \times \frac{22,400 \text{ cc gas}}{6.023 \times 10^{23} \text{ atom}} = \underline{\underline{108 \text{ cc gas}}}$$

For Powder

$$1.96 \times 10^{19} \text{ } 132 \text{ cc gas}$$

Calculation of % gas released:

For Pellets

$$\frac{108}{108} = 100\% \text{ gas release}^*$$

For Powder

$$\frac{132}{108} = 122\% \text{ gas release}^*$$

*These numbers may result from erroneous Kr-85 gamma counting results.

APPENDIX C

METALLOGRAPHY AND AUTORADIOGRAPHY
RESULTS

C-1 PELLET FUEL, SECTION A, FIGURE C-1

Linear Power	20.4 kW/ft
Fuel Center Temperature	2600°C
Fuel Surface Temperature	1110°C
Cladding Inside Surface Temperature	660°C (1220°F)
Void Volume	3%

The central void occupied only 3% of the cross-sectional area in this specimen. Since this void size is inconsistent with the fuel densification as indicated by the columnar grain radius (80%), it was concluded that molten fuel from the upper section relocated to this region. The overall fuel morphology was similar to pellet fuel section E (5.2.2) which ran at 22 kW/ft.

With the exception of one isolated region of 0.002 in. no significant fuel-cladding chemical interaction was observed. Cladding inside surface temperatures in this region were calculated to be 1220°F compared to 1330°F for specimen E. Autoradiographs are presented in Figure C-9. No distinctive characteristics were noted.

C-2 PELLET FUEL, SECTION H, FIGURE C-2

Linear Power	23.5 kW/ft
Fuel Center Temperature	2600°C
Fuel Surface Temperature	1260°C
Cladding Inside Surface Temperature	760°C (1400°F)
Void Volume	28%

This specimen represents the peak power section of the pellet fuel where the central void occupied 28% of the fuel volume. This void size is inconsistent with the initial fuel density and it is reasonable to consider that the fuel melted to the radius of the center void and subsequently moved to the bottom area of the pin. A thin band of fuel exhibiting the characteristic dendritic microstructure associated with molten fuel was visible and covered about 20% of the inside surface of the center void (see Figure C-2).

The fuel area adjacent to the cladding contained large quantities of metallic material, extending in several places as wedge-shaped sections well into the fuel. A radial crack in the fuel which extended to the central void was associated with each of these wedges. This metallic material was attacked by an electrolytic etch in a manner similar to the cladding and exhibited neither alpha or beta-gamma activity on autoradiographs, Figure C-8. A minimum cladding thickness of 0.0116 inch was measured in this section, indicating a loss of about 0.004 inch of cladding material. Cladding inside surface temperature of 1400°F was calculated.

C-3 POWDER FUEL, SECTION B, FIGURE C-3

Linear Power	23.3 kW/ft
Fuel Center Temperature	Molten
Fuel Surface Temperature	1260°C
Cladding Inside Surface Temperature	760°C (1400°F)
Void Volume	0

This specimen represents the peak power section of the powder fuel and is equivalent to section H of the pellet fuel, (see Figure C-2). Since the bottom end of the powder fuel ran at highest power, fuel relocated into rather than away from this area. The central fuel exhibited a dendritic structure and was surrounded by a pore-free ring typical of molten fuel. A 100X radial micrograph is shown in Figure C-4, with a 500X section showing fuel-cladding reaction. A minimum cladding thickness of 0.0119 inch was measured exclusive of the affected zone. Autoradiographs are presented in Figure C-8. Decreased beta-gamma activity was noted in the dendritic central region.

C-4 POWDER FUEL, SECTION A, FIGURE C-5

The top of this specimen is the opposite face of the transverse sample described in Appendix C-3. Typical fuel-cladding interaction was observed at the top area of this section. The character of this reaction changed from severe to insignificant over an axial distance equivalent to about one cladding thickness (see designated area in Figure C-5). Since the fuel structure showed no significant change in this area, it was concluded that end cooling reduced the cladding temperature below the reaction threshold. At the lower end of the fuel column gross diffusion of the mixed oxide into the UO_2 powder insulator was observed. The full extent of this mixing may be seen in Figure C-6, where autoradiographs delineate fission product and fuel activity. The loop of fused material through the insulator powder is not easily explained. Recent microprobe examinations of material with similar appearance indicates that it may be cesium uranate.

C-5 AUTORADIOGRAPHY

Alpha and beta-gamma autoradiographs of the fuel sections are shown in Figures C-6 through C-9. In addition to the specimens previously discussed there is included a longitudinal specimen which illustrates the bridges found across the central void of the pellet fuel (subsection G). This bridge had enhanced beta-gamma activity suggesting a high concentration of fission products. Axial gamma scans also indicated the location of these bridges.

An attempt was made to expose all autoradiographs for the same time period. Since the time was only 2 seconds, this was difficult and some variation in exposure time occurred.

The exposure of highly enriched fuel in a thermal flux resulted in severe surface-to-center flux depression in the fuel. The high activity levels found in the outer fuel were therefore expected. Fission-product migration may have enhanced this phenomenon.

30X

LINEAR POWER	20.4 kW/ft
FUEL CENTER TEMPERATURE	2600°C
FUEL SURFACE TEMPERATURE	1110°C
CLAD INSIDE SURFACE TEMPERATURE	660°C (1220°F)
VOID VOLUME	3%

Figure C-1 Pellet Fuel Section A

30X

LINEAR POWER	23.5 kW/ft
FUEL CENTER TEMPERATURE	2600°C
FUEL SURFACE TEMPERATURE	1260°C
CLAD INSIDE SURFACE TEMPERATURE	760°C (1400°F)
VOID VOLUME	28%

Figure C-2 Pellet Fuel Section H

30X

LINEAR POWER	23.5 kW/ft
FUEL CENTER TEMPERATURE	MOLTEN
FUEL SURFACE TEMPERATURE	1260°C
CLAD INSIDE SURFACE TEMPERATURE	760°C (1400°F)
VOID VOLUME	0

Figure C-3 Powder Fuel Section B

0.005 in.

Photos reduced to 74%
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0.001 in.

AS POLISHED RADIAL SECTION 100X

FUEL-CLAD INTERACTION 500X

Figure C-4 Powder Fuel Section B Micrographs

Figure C-5 Powder Fuel Section A

α AUTORADIOGRAPH

β - γ AUTORADIOGRAPH

MICROGRAPH

10X

Photos reduced to 95%
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Figure C-6 Powder Fuel Section A Autoradiography

PELLET FUEL SECTION 10X

POWDER FUEL SECTION 10X

β - γ AUTORADIOGRAPH

β - γ AUTORADIOGRAPH

α AUTORADIOGRAPH

α AUTORADIOGRAPH

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Figure C-7 Pellet Fuel Section E and Powder Fuel Section E Autoradiography

PELLET FUEL SECTION H 10X

POWDER FUEL SECTION B 10X

β - γ AUTORADIOGRAPH

β - γ AUTORADIOGRAPH

α AUTORADIOGRAPH

α AUTORADIOGRAPH

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Figure C-8 Powder Fuel Section B and Pellet Fuel Section H Autoradiography

PELLET FUEL SECTION G 10X

PELLET FUEL SECTION A 10X

$\beta - \gamma$ AUTORADIOGRAPH

$\beta - \gamma$ AUTORADIOGRAPH

α AUTORADIOGRAPH

α AUTORADIOGRAPH

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Figure C-9 Pellet Fuel Section A and Pellet Fuel Section G Autoradiography

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Engineering Center
Atoms International
P. O. Box 1449
Canoga Park, California 91304

Manager—Advanced Development Activity
General Electric Company
Knolls Atomic Power Laboratory
P. O. Box 1072
Schenectady, New York 12301

Director, LMFBR Technology Program
Atoms International
P. O. Box 309
Canoga Park, California 91304

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Nuclear Development Center
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1	Manager Liquid Metal Information Center Atoms International P. O. Box 1449 Canoga Park, California 91304	1
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