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**EXPERIMENTAL STUDIES OF
TRANSIENT EFFECTS IN FAST REACTOR FUELS
SERIES III,
PRE-IRRADIATED MIXED OXIDE ($\text{PuO}_2\text{-UO}_2$)
IRRADIATIONS FINAL REPORT,
TRANSIENT IRRADIATIONS**

J.E. HANSON
J.H. FIELD

U.S. ATOMIC ENERGY COMMISSION
CONTRACT AT(04-3)-189
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ADVANCED PRODUCTS OPERATION

GENERAL ELECTRIC

SUNNYVALE, CALIFORNIA

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FINAL REPORT, TRANSIENT IRRADIATIONS

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TABLE OF CONTENTS

		<u>Page</u>
	ABSTRACT	1
SECTION I	SUMMARY AND CONCLUSIONS	1
	1.1 Summary	1
	1.2 Conclusions	2
SECTION II	INTRODUCTION	2
SECTION III	RESULTS	3
	3.1 Transient Irradiations	4
	3.2 Post-Irradiation Examinations	9
SECTION IV	DISCUSSION OF RESULTS	95
	4.1 Derivation of Transient Fission Gas Release Model from the C3B Experiment Results	95
	4.2 Transient Performance of Irradiated Fuel with Respect to a Postulated Credible Accident in an FCR (Specimen C3E)	104
	4.3 Transient Performance Limit of Highly Irradiated Fuel (Specimen C3C)	107
	4.4 Thermal versus Fast Flux Irradiation Performance	109
	4.5 Fission Product Swelling in High Burnup Fuel	112
APPENDIX A	FUEL SPECIMEN AND CAPSULE DETAILS	113
APPENDIX B	COMPARISON OF MEASURED FISSION GAS RELEASE TO CALCULATED FISSION GAS RELEASE FOR SPECIMEN C3A	127
APPENDIX C	ESTIMATE OF FISSION GAS RELEASE FROM SPECIMEN C3E	131
APPENDIX D	ESTIMATION OF INTERNAL PRESSURES IN SPECIMEN C3C	147
APPENDIX E	CALCULATION OF FUEL SPECIMEN TEMPERATURES	153
	ACKNOWLEDGMENTS	163
	REFERENCES	163
	DISTRIBUTION	165

LIST OF ILLUSTRATIONS

<u>Figure</u>	<u>Title</u>	<u>Page</u>
3-1	Transient Irradiation Data, Transient C3B-1	6
3-2	Calculated Temperatures—Specimen C3B, Transient C3B-2	7
3-3	Calculated Radial Temperature Profiles in Specimen C3B, Transient C3B-2	8
3-4	Transient Irradiation Data, Transient C3E-1	10
3-5	Calculated Radial Temperature Profile in Specimen C3E for Maximum Molten Fuel Radius ($t = 4.4$ sec)	11
3-6	Transient Irradiation Data, Transient C3C-1	12
3-7	Calculated Radial Temperature Profile in Specimen C3C for Maximum Molten Fuel Radius ($t = 2.9$ sec)	13
3-8	Neutron Radiograph and Fuel Pin Schematic Specimen C3E (63,000 MWd/Te Burnup)	14
3-9	Neutron Radiograph and Fuel Pin Schematic Specimen C3C (64,000 MWd/Te)	15
3-10	Comparison of C3B Gamma Scans Before and After Transient Irradiation	16
3-11	Profile Measurements Before and After Irradiation, Specimen C3B	17
3-12	Specimen C3B Following Transient Irradiation	18
3-13	Longitudinal Section from Top of Specimen C3B Following Transient Irradiation	19
3-14	Photomicrograph of Fuel-Cladding Interface in Top Longitudinal Section, Specimen C3B	22
3-15	Longitudinal Section from Lower Half of Specimen C3B Following Transient Irradiation	23
3-16	Longitudinal Section from Bottom of Fuel Column of Specimen C3B Following Transient Irradiation	25
3-17	Transverse Section from Bottom of Fuel Column, Specimen C3B	27
3-18	Diametral Composite of Transverse Section from Bottom of Specimen C3B Fuel Column	29
3-19	Transverse Section from Upper Portion of Fuel Specimen C3B	32
3-20	Diametral Composite of Transverse Section from Top Portion of Specimen C3B Fuel Column	33
3-21	High Magnification Photomicrograph Illustrating Porosity (Fission Gas Bubbles) Trails and Grain Growth	35
3-22	High Magnification Photomicrographs from Regions of Fuel Indicated in Figure 3-18	39
3-23	Photomicrographs Illustrating Cracking and Grain Boundary Corrosion in Cladding from Bottom Longitudinal Section of Specimen C3B	41
3-24	Photomicrographs Illustrating Fuel-Cladding Reactions and Cladding Cracking in Specimen C3B	43
3-25	Metallographic Examination of Unirradiated Type 347 Stainless Steel Cladding	45
3-26	Specimen C3A Cladding Structure Following Steady-State Irradiation	46
3-27	Post-Steady-State-Irradiation Gamma Scan of Capsule C3E	47
3-28	Post-Transient Irradiation Gamma Scans of Capsule C3E	49
3-29	Specimen C3E Following Steady-State and Transient Irradiation	51
3-30	Photomacrograph and Beta-Gamma and Alpha Autoradiographs of Longitudinal Section from Bottom of Specimen C3E	53

LIST OF ILLUSTRATIONS (Continued)

<u>Figure</u>	<u>Title</u>	<u>Page</u>
3-31	Photomacrograph and Beta-Gamma and Alpha Autoradiographs of Longitudinal Section from Center of Specimen C3E	55
3-32	Photomacrograph and Beta-Gamma and Alpha Autoradiographs of Longitudinal Section from Top of Specimen C3E	57
3-33	Photomacrograph and Beta-Gamma and Alpha Autoradiographs of Transverse Section from Center of Specimen C3E	59
3-34	Photomacrograph and Beta-Gamma and Alpha Autoradiographs of Transverse Section from Top Portion of Specimen C3E	60
3-35	Photomicrograph Illustrating Molten Boundary Radius and Distribution of Metallic Fission Products in Longitudinal Section Shown in Figure 3-30	61
3-36	Photomicrographs Illustrating Molten Boundary Radius and Distribution of Metallic Fission Products in Longitudinal Section Shown in Figure 3-31	63
3-37	Photomicrographs of Area of Relocated Fuel Shown in Figure 3-32 (Specimen C3E)	65
3-38	Composite Photomicrograph and High Magnification Photomicrographs of Transverse Cross Section Shown in Figure 3-33 (Specimen C3E, As-Polished)	67
3-39	Composite Photomicrograph and High Magnification Photomicrographs of Transverse Cross Section Shown in Figure 3-33 (Specimen C3E, Etched)	69
3-40	Composite Photomicrograph and High Magnification Photomicrographs of Transverse Cross Section Shown in Figure 3-34 (Specimen C3E, As-Polished)	71
3-41	Composite Photomicrograph and High Magnification Photomicrographs of Transverse Cross Section Shown in Figure 3-34 (Specimen C3E, Etched)	73
3-42	Metallographic Examination of Specimen C3E Cladding (347 SS)	75
3-43	Gamma Scans of Capsule C3C	77
3-44	Fuel Redistribution Following Transient Irradiation of Specimen C3C	79
3-45	Fuel Pin Exterior and As-Cut Sections from Specimen C3C	81
3-46	Longitudinal and Transverse Sections from Specimen C3C	83
3-47	Photomacrograph and Alpha and Beta-Gamma Autoradiographs from Longitudinal Section from Specimen C3C	85
3-48	Photomicrographs of Fuel and Cladding from Longitudinal Section of Specimen C3C Shown in Figure 3-47	87
3-49	Photomacrographs, Photomicrographs, and Alpha and Beta-Gamma Autoradiographs from Transverse Section of Specimen C3C Shown in Figure 3-46e	89
3-50	Photomacrograph, Photomicrographs, and Alpha and Beta-Gamma Autoradiographs of Transverse Section from Specimen C3C Shown in Figure 3-46f	91
3-51	Photomacrograph, Photomicrographs, and Alpha and Beta-Gamma Autoradiographs from Transverse Section from Specimen C3C Shown in Figure 3-46g	93
4-1	Calculated Power Profile in Specimen C3B	96
4-2	Fission Gas Generation in Specimen C3B	97
4-3	Kdt and K versus Temperature	99
4-4	Calculated Steady-State Temperature Profile in Specimen C3B, $Q' = 18.1 \text{ kW/ft}$	100

LIST OF ILLUSTRATIONS (Continued)

<u>Figure</u>	<u>Title</u>	<u>Page</u>
4-5	Steady-State Temperature Profile and Fission Gas Generation, Specimen C3B	101
4-6	Stress-Strain Diagram for Type 347 Stainless Steel at 1400°F	105
4-7	Stress-Strain Diagram for Type 347 Stainless Steel at ~1850°F	106
4-8	Comparison of Transient Experiment Results with Calculated Fast Ceramic Reactor Temperature Profile during \$0.20/sec Excursion	108
4-9	Gas Generation in Fast and Thermal Fluxes	110
4-10	Fission Gas Release in Reference FCR Fuel	111
A-1	Capsule Assembly	115
A-2	TREAT Capsule Assembly for Transient Irradiation of Pre-Irradiated Specimens	117
A-3	Fuel Specimen Details	119
A-4	Pressure Transducer Installation, TREAT Series III Capsules	121
A-5	Steps in the Remote Installation of Pressure Transducer in Specimen C3B	123
B-1	Specimen C3A Radial Temperature Profile and Fission Gas Generation	128
C-1	Calculated Power Profile in Specimen C3E	132
C-2	Calculated Steady-State Temperature Profile in Specimen C3E at End of Irradiation	133
C-3	Fission Gas Generation in Specimen C3E	134
C-4	Steady-State Temperature Profile and Fission Gas Generation, Specimen C3E	137
C-5	GETR Pool Position Axial Flux Shapes	138
C-6	Microcore Samples from Specimen C3E	143
D-1	Calculated Power Profile in Specimen C3C	149
D-2	Calculated Steady-State Temperature Profile in Specimen C3C at End of Irradiation	150
D-3	Fission Gas Generation in Specimen C3C	150
D-4	Yield Strength versus Temperature in Type 347 Stainless Steel	152
E-1	Geometric Model for Argus Calculations	153
E-2	Thermal Conductivity of PuO ₂ -UO ₂ Fuel	154
E-3	ρC_p of Mixed Oxide Fuel	155
E-4	Thermal Conductivity for Type 347 Stainless Steel (Annealed)	156
E-5	ρC_p for Type 347 Stainless Steel	157
E-6	Thermal Conductivity of NaK (78 Percent K, 22 Percent Na)	158
E-7	ρC_p of NaK	159
E-8	Thermal Conductivity of 6061 Aluminum	160
E-9	ρC_p of 6061 Aluminum	161

LIST OF TABLES

<u>Table</u>	<u>Title</u>	<u>Page</u>
2-1	Summary of Test Objectives in Series III Transient Irradiations	3
3-1	Steady-State Irradiation History of Series III Capsules Based on Thermocouple Data	5
3-2	TREAT Reactor Parameters for Series III Transients	5
3-3	Compilation of Results from Test Transients	5
3-4	Dimensional Measurements Specimen C3B	13
3-5	Gas Composition and Volume in Specimen C3E	38
3-6	Isotopic Analysis of Fission Gas in Specimen C3E	38
3-7	Specimen C3E Burnup Data	76
3-8	Specimen C3C Diameter Measurements	80
A-1	Data for Series 3 Fuel Pins	120
A-2	Isotopic Analysis of Plutonium Feed Solution for Series III for Fuel Pellets	120
A-3	Pressing and Sintering Data for Series III Pellets	125
C-1	Conversion of Xe-135 to Xe-136 in FCR Fuel Specimen Thermal Flux Irradiations	140
C-2	Xenon and Krypton Yields for Thermal Fission of U-235 and Pu-239	140
C-3	Alpha Radiograph Structure and Micro-Core Location	144
C-4	Plutonium Isotopic Abundances	144
C-5	Alpha Pulse Height Analysis	144
C-6	Plutonium-to-Uranium Ratio	145
C-7	Relative Alpha Specific Activities	145
C-8	Radial Distribution of Ce-144 and Cs-137	145

PREFACE

This report presents a compilation of material previously described in periodical reports under the Fast Ceramic Reactor Development Program. For a chronological record of this work, the reader is referred to the following reports:

Fourteenth Quarterly Report, GEAP-4860, May 1965
 Fifteenth Quarterly Report, GEAP-4931, August 1965
 Sixteenth Quarterly Report, GEAP-4982, January 1966
 Seventeenth Quarterly Report, GEAP-5098, February 1966
 Eighteenth Quarterly Report, GEAP-5158, August 1966

In addition, a substantial further body of work has accrued in this area since the conclusion of the effort reported here, and in some cases may substantially qualify the results of that effort. Specifically, experiments involving fast flux pre-irradiation have been initiated and transient results as a function of axial restraint, smeared density, and fuel form (pellet versus powder) have been reported in the following reports:

Nineteenth Quarterly Report, GEAP-5198, August 1966
 Twentieth Quarterly Report, GEAP-5292, November 1966
 Twenty-First Quarterly Report, GEAP-5433, February 1967
 Twenty-Second Quarterly Report, GEAP-5491, May 1967
 Twenty-Third Quarterly Report, GEAP-5522, August 1967

and an ANS paper, Thomas, G. R., and Field, J. H., "Response of Axially-Restrained Pellet Fuel to Transient Overpower," presented at the ANS Winter Meeting, November 5—9, 1967, Chicago, Illinois.

ABSTRACT

Presented in this report are the results of an experimental program to evaluate the performance of pre-irradiated fast ceramic reactor fuel under conditions simulating accidental power excursions. Results from three experiments in the TREAT facility using 0.250-inch-o.d., Type-347 stainless steel cladding, 28 weight percent PuO_2 - 72 weight percent UO_2 fuel specimens are interpreted with regard to fuel and cladding temperatures, fuel relocation, fission gas release, and clad-

ding integrity. The transient failure threshold for axially unrestrained fuel at $\sim 70,000$ MWd/Te* was identified at ~ 50 volume percent molten fuel. The primary mechanism for this transient failure was identified as internal pressure caused by the sudden local release of contained fission gas during the excursion.

*Megawatt-day per metric tonne.

SECTION I

SUMMARY AND CONCLUSIONS

1.1 SUMMARY

Transient irradiations of three pre-irradiated mixed oxide, axially unrestrained fuel specimens were conducted in the TREAT Facility at the National Reactor Testing Station (NRTS) in Idaho. The three specimens, designated C3B, C3C, and C3E, were irradiated at steady-state conditions in the General Electric Test Reactor (GETR) to burnups of 8,100, 73,000, and 70,000 MWd/Te,* respectively. Neutron radiography of the high burnup specimens (C3C and C3E) following completion of the steady-state irradiation disclosed the presence of a center void, some axial fuel relocation, axial elongation of the fuel column, and some evidence of possible plutonium segregation in each specimen. No evidence of cladding expansion was found in either case.

1.1.1 Specimen C3B

Transient irradiation of the low burnup specimen (8,100 MWd/Te) essentially duplicated conditions of fuel and cladding temperature obtained in the transient irradiation of a zero burnup specimen (C2B).⁽⁴⁾ The results obtained in the C3B experiment were substantially different from those obtained in

the C2B experiment, however. In addition to cladding deformation over the fueled length and cracking of the cladding at the inside surface (these effects did not occur in the C2B experiment), the axial fuel relocation pattern in the C3B experiment suggested the expansion of a fission gas bubble, rather than gravity, as the principal driving force for fuel relocation. In the C3B experiment, axial fuel relocation occurred in both the upward and downward direction (fission gas bubble) whereas, in the C2B experiment, fuel relocation occurred only in the downward direction (gravity). A model for fission gas release during a transient was formulated on the basis of the results of this experiment.

1.1.2 Specimen C3E

The first transient irradiation of a high burnup specimen (C3E, 70,000 MWd/Te) resulted in conditions of cladding and fuel temperature somewhat more severe than those calculated to occur in postulated credible accidents in a Fast Ceramic Reactor (FCR). In this experiment no cladding deformation or significant fuel relocation occurred. This was as predicted by application of the model derived from interpretation and analysis of the results from the C3B experiment. The formation of a metallic fission product ingot,

*Based on 200 MeV/fission and Te of Pu + U (metal).

apparent plutonium segregation, and graphic evidence of the molten boundary radius in oxide fuel were observed during the post-irradiation examination of this specimen.

1.1.3 Specimen C3C

Transient irradiation of the second high burnup specimen (C3C, 73,000 MWd/Te) served to substantiate further the model for gas release during a transient and to establish a transient performance limit for irradiated oxide fuel. Application of the model predicted cladding deformation of about 1 percent with a molten boundary radius of 0.090 inch (67 volume percent). Post-irradiation examination disclosed a molten boundary of 0.090 inch and 0.91 percent cladding deformation. Cracking, and a heavy grain boundary precipitate at the inner surface of the cladding which appeared identical to that in the low burnup specimen were also observed. Failure of the diaphragm at the bottom of the fuel pin resulted in the expulsion of the molten fuel into the bottom of the capsule, which demonstrated the fluidity of the molten fuel and the substantial driving force available from the release of fission gases.

1.2 CONCLUSIONS

Interpretation and analysis of the results obtained from the three experiments have led to the following conclusions:

- a. The release of fission gases from fuel which is melted during a transient can be one of the primary mechanisms for cladding failure under these conditions.
- b. The release of fission gases from fuel which is melted during a transient provides a substantial driving force for redistribution and dispersion of the fuel.
- c. Under the conditions of these tests, cladding deformation of about 1 percent will result in gross cracking at the inner surface of the cladding due to the formation of heavy grain boundary precipitation in this region.
- d. From these tests it appears that foaming of irradiated oxide fuel does not occur even when a substantial portion of the fuel is melted.
- e. On the basis of these experiments there appears to be considerable margin between the demonstrated performance limit (~1 percent cladding deformation and >50 percent molten fuel) and postulated credible accidents in an FCR inasmuch as current design criteria and safety analyses do not result in any fuel becoming completely molten during the postulated credible accidents.

SECTION II

INTRODUCTION

This is the third report of a series concerning the irradiation of oxide fuels under transient conditions which simulate the effects of fast reactor power excursions. Presented in the first report⁽¹⁾ were the results and

conclusions from transient irradiations of large-diameter (1-inch) UO_2 fuel rods. This work (Series I irradiations) was primarily directed toward fuel development for the Experimental Fast Ceramic Reactor⁽²⁾ which

is now known as SEFOR.⁽³⁾ The results and conclusions from transient irradiations of three 0.250-inch-diameter, zero burnup, mixed oxide ($\text{PuO}_2\text{-UO}_2$) fuel rods were presented in the second report.⁽⁴⁾ This work (Series II irradiations), as well as the work to be reported herein, was directed toward the development of mixed oxide fuel for a large FCR.^(5, 6, 7)

This program of fuel capsule transient irradiations in the TREAT facility⁽⁸⁾ is based to a large extent on a prior analytical study of transient effects in fast reactor fuels.⁽⁹⁾ Transient irradiations of zero burnup mixed oxide fuel (Series II irradiations) were designed to investigate fuel-cladding transient performance with regard to:

- a. Densification and redistribution of the fuel as a result of transient melting.
- b. Cladding deformation and possible failure from high thermal and pressure stress arising from very high heat fluxes and possible fuel vapor pressure.

Transient irradiations of pre-irradiated mixed oxide fuel, the subject of this report, were designed to investigate the added effects on fuel-cladding transient performance of fission product formation, fission gas release, fuel relocation and densification (central void formation), cladding damage, and changes in thermal properties resulting from short- and long-term steady-state irradiation.

The data and conclusions presented in this report are based on the results of four transients conducted on three pre-irradiated, 0.250-inch-diameter mixed oxide fuel specimens. Since the objective of this series of tests was to define incipient fuel-cladding failure for irradiated fuel, the approach taken was to follow a transient irradiate-examine sequence prior to proceeding to the transient irradiation of the next specimen. The objectives of each test in this series are summarized in Table 2-1.

TABLE 2-1

SUMMARY OF TEST OBJECTIVES IN
SERIES III TRANSIENT IRRADIATIONS

<u>Specimen</u>	<u>Goal Burnup* (MWd/Te)</u>	<u>Test Objective</u>
C3B	8,000	Compare transient fuel-cladding performance to previous zero burnup specimen (C2B)
C3E	65,000	Investigate fuel-cladding performance of high burnup specimen under calculated credible FCR accident conditions
C3C	65,000	Investigate fuel-cladding performance of high burnup specimen subjected to gross center melting of the fuel during a transient.

*Based on 200 MeV/fission and Te of Pu+U.

SECTION III

RESULTS

In the previous test series, three 0.250-inch-diameter, zero burnup, mixed oxide fuel specimens (designated C2A, C2B, and C2C) were transient irradiated in the TREAT facility.

The results and conclusions from this series of tests (Series II irradiations) were presented in Reference (4).

The test series discussed herein involved five 0.250-inch-diameter, mixed oxide fuel specimens (designated C3A, C3B, C3C, C3D, and C3E) which were steady-state irradiated in pool positions in the GETR to goal exposures of about 8,000 MWd/Te (specimens C3A and C3B) and 65,000 MWd/Te (specimens C3C, C3D, and C3E).

Two of these capsules, C3A and C3D, were destructively examined in the Radioactive Materials Laboratory (RML) of the Vallecitos Nuclear Center. The three remaining capsules were nondestructively examined prior to remote reencapsulation for transient irradiation in the TREAT facility. The steady-state irradiation histories and post-irradiation destructive and nondestructive examinations of capsules C3A, C3B, C3C, C3D, and C3E were presented and discussed in an interim report.⁽¹⁰⁾ A resume of the steady-state irradiation histories of the five Series III capsules is given in Table 3-1.

Throughout this report, reference will be made to GEAP-4819⁽¹⁰⁾ as the "interim report," and it will be assumed that the reader has this companion document available.

3.1 TRANSIENT IRRADIATIONS

The Series III transient irradiation experiments involved four transients on three fuel specimens. One of these transients, the first on specimen C3B, was a relatively small transient for calibration purposes. The remaining three were the irradiations which are considered the test transients. The TREAT reactor parameters for each transient are given in Table 3-2, and a compilation of pertinent specimen performance data is given in Table 3-3.

3.1.1 Specimen C3B

As seen in Table 3-2, specimen C3B was subjected to two clipped transients. The

first, of 28.4-MW peak power, was conducted to verify physics and heat transfer data for the low burnup specimen. The data obtained from this transient (C3B-1) are illustrated in Figure 3-1. As can be seen, the agreement between measured temperatures and calculated temperatures was quite good, thus corroborating the physics and heat transfer calculations.

On the basis of the experimentally verified physics and heat transfer data, a second transient, C3B-2, was run. As indicated in Table 2-1, the objective of this test was to compare the transient performance of this low burnup specimen to that of a zero burnup specimen [C2B⁽⁴⁾] which attained a peak fuel temperature of 5000°F and a maximum cladding temperature of 1800°F. Prior to installing the capsule in the TREAT reactor for the second transient, a routine instrument check disclosed that all of the internal thermocouples had failed by open-circuiting. Thus, the data presented in Figure 3-2 are based solely upon transient heat transfer calculations and post-irradiation examination of the fuel. Depicted in Figure 3-3 are calculated radial temperature profiles in the fuel at various times during the transient. Of particular interest is the calculated molten boundary radius. As will be discussed in greater detail later, there appears to be good agreement between the calculated molten boundary radius and the molten boundary radius seen in autoradiographs and photomicrographs of the fuel.

3.1.2 Specimen C3E

As will be discussed in greater detail in a following section, the results obtained in the C3B-2 transient were sufficiently different than those anticipated, as to require that the transient irradiation of the first high burnup specimen be as typical as possible of postulated credible accident conditions in an FCR. Accordingly, the basis for the design of transient C3E-1 was a pulse which would result in a maximum cladding temperature of 1400°F, a

TABLE 3-1
STEADY-STATE IRRADIATION HISTORY OF SERIES III
CAPSULES BASED ON THERMOCOUPLE DATA

GETR Cycle Number	Peak Rod Power (kW/ft)					Average Rod Power (kW/ft)					Burnup (MWd/Te)*				
	Capsule					Capsule					Capsule				
	C3A	C3B	C3C	C3D	C3E	C3A	C3B	C3C	C3D	C3E	C3A	C3B	C3C	C3D	C3E
42	29.3	25.1	--	--	--	20.4	18.1	--	--	--	9,200	8,100	--	--	--
44	--	--	27.9	28.8	30.8	--	--	19.7	19.7	20.5	--	--	7,100	7,100	7,300
45	--	--	27.7	28.6	24.0	--	--	21.9	22.4	17.6	--	--	8,100	8,400	6,500
46	--	--	26.7	24.3	25.8	--	--	19.2	19.2	18.2	--	--	7,200	7,200	6,800
47	--	--	27.3	29.2	23.9	--	--	20.4	19.9	17.8	--	--	6,700	6,600	5,900
48	--	--	27.5	26.0	26.0	--	--	20.1	20.9	20.1	--	--	7,600	7,900	7,600
49	--	--	27.9	31.7	23.1	--	--	19.6	22.1	16.9	--	--	9,000	10,200	7,800
50	--	--	26.9	27.5	22.3	--	--	19.9	21.9	18.1	--	--	8,600	9,500	7,800
51	--	--	25.1	24.9	22.2	--	--	18.6	19.3	17.7	--	--	6,100	6,300	5,700
52	--	--	23.1	22.2	22.2	--	--	18.6	17.8	18.7	--	--	6,400	6,200	6,500
53	--	--	25.3	25.0	23.9	--	--	17.0	17.6	20.2	--	--	6,500	6,800	7,800
													73,000	76,000	70,000

*MWd/Te = megawatt days per metric tonne of Pu + U at 200 MeV/fission. Peak values.
Average over 6-inch fuel length is ~ 1.5 percent less.

TABLE 3-2
TREAT REACTOR PARAMETERS FOR SERIES III TRANSIENTS

APO Transient Number	TREAT Transient Number	Initial Period (sec)	Reactivity Addition (percent Δk)	Integrated Power (MW-sec)	Peak Power (MW)	Type of Transient
C3B-1	822	0.622	0.753	94	28.4	Clipped after peak power.
C3B-2	834	0.217	1.08	193	149	Clipped after peak power.
C3E-1	926	0.239	1.04	160.5	124	Clipped after peak power.
C3C-1	982	0.157	1.25	239	273	Clipped after peak power.

TABLE 3-3
COMPILATION OF RESULTS FROM TEST TRANSIENTS

Specimen	Reactor Power		Sample Energy Release (Btu)	Calculated Temperatures		Post-Irradiation Condition
	Peak (MW)	Integrated (MW-sec)		Maximum Fuel Temperature (°F)	Maximum Average Cladding (°F)	
C3B	149	193	68	5280	1830 (800°F preheat)	Cladding deformation and cracking, axial separation of the fuel column.
C3E	124	160.5	44	4900	1245 (600°F preheat)	No radial deformation or cracking of cladding. Some axial movement of the fuel.
C3C	273	239	67	5950	1295 (300°F preheat)	Cladding deformation and cracking. Axial expulsion of molten fuel through bottom of fuel pin.

FIGURE 3-1. TRANSIENT IRRADIATION DATA, TRANSIENT C3B-1

FIGURE 3-2. CALCULATED TEMPERATURES - SPECIMEN C3B, TRANSIENT C3B-2

FIGURE 3-3. CALCULATED RADIAL TEMPERATURE PROFILES IN SPECIMEN C3B, TRANSIENT C3B-2

maximum fuel temperature of 4900°F, and a maximum of 30 volume percent molten fuel, since these conditions represent maximum fuel and cladding temperatures calculated to occur in a credible excursion accident in an FCR.⁽¹¹⁾

The results attained, as illustrated in Figure 3-4, were quite close to the target values, with a maximum fuel temperature of 4900°F, and a maximum average cladding temperature of about 1300°F. A calibration transient was not run on this sample since it was deemed most important to have functioning thermocouples during the test transient (it will be recalled that the thermocouples in capsule C3B failed after the calibration transient). The calculated radial temperature profiles in the fuel and cladding at various times during the transient are illustrated in Figure 3-5.

Post-irradiation examination of specimen C3E disclosed that no significant damage occurred to the fuel specimen as a result of the test transient.

To establish a transient performance limit for high burnup mixed oxide fuel, transient C3C-1 was based on the objective of melting sufficient fuel to release enough fission gas to cause approximately 1 percent cladding deformation. Attainment of these experimental objectives would verify the following:

- a. The validity of the model derived from the C3B experiment.
- b. The apparent cladding brittleness which was observed in the C3B experiment where severe cladding cracking occurred at ~1 percent transient deformation.

The thermocouple data and calculated temperatures for transient C3C-1 are presented in Figure 3-6, the calculated radial temperature profiles in the fuel and cladding during the transient are presented in Figure 3-7.

3.2 POST-IRRADIATION EXAMINATIONS

3.2.1 Neutron Radiography (Specimens C3E and C3C)

Following the completion of the preparation of the interim report⁽¹⁰⁾ on the Series III irradiations, it was learned that neutron radiograph facilities were available at Battelle Memorial Institute. Accordingly, arrangements were made to neutron radiograph the two high burnup specimens, C3C and C3E. The radiographs obtained, Figures 3-8 and 3-9, disclosed that cladding failure had not occurred in these specimens. In addition, the radiographs established the condition of the fuel with regard to fuel column elongation, axial fuel redistribution, and center void formation, prior to transient irradiation. Ramifications of the observed fuel column elongation will be discussed in greater detail at a later point in this report.

3.2.2 Specimen C3B Post-Irradiation Examination

Post steady-state irradiation examination of specimen C3A⁽¹⁰⁾ established the condition of the fuel in specimen C3B prior to transient irradiation since these two specimens were steady-state irradiated at near identical conditions (see Table 3-1).

The radioactivity resulting from the steady-state pre-irradiation of this specimen necessitated remote disassembly. Removal of the inner capsule assembly from the outer capsule assembly (see Figures A-1 and A-2, Appendix A) was accomplished in the RML gamma cells. Disassembly of the inner capsule assembly was accomplished in the RML alpha enclosure. Prior to disassembly in the alpha enclosure, the capsule was gamma scanned. A comparison of the gamma scans prior to, and following, transient irradiation is given in Figures 3-10(a) and 3-10(b), respectively. In view of the change in the pattern of the scans,

FIGURE 3-4. TRANSIENT IRRADIATION DATA, TRANSIENT C3E-1

FIGURE 3-5. CALCULATED RADIAL TEMPERATURE PROFILE IN SPECIMEN C3E FOR MAXIMUM MOLTEN FUEL RADIUS ($t = 4.4$ sec)

it was concluded that fuel movement had occurred during the transient.

As the first step in the disassembly of the capsule, the capsule wall was punctured and the NaK cover gas was sampled for fission gases. The detection of fission gases indicated that failure of the fuel specimen cladding had occurred. An attempt to analyze the gas sample obtained was less than successful since the majority of the gas (>98 percent) was helium which was used as the NaK cover gas.

As seen in the exterior view of the pin, there was no evidence of gross failure of the cladding. Measurement of the fuel pin diameter, however, disclosed that some radial deformation of the pin had occurred. The dimensional measurements for specimen C3B are presented in Table 3-4. As is evident from Table 3-4, approximately 2 to 3 mils radial deformation over the fueled portion of the pin was measured. Defor-

mation of this nature was one of the more surprising results of this test.

It is illustrated in Figure 3-11 that bowing of the radially unrestrained fuel pin was also quite pronounced.

Sectioning of the fuel pin revealed that fuel movement had, indeed, occurred. This is evidenced in Figure 3-12 by the discontinuous center void in the as-cut sections and the lack of a center void in the top and bottom longitudinal sections.* Closer study of the longitudinal sections from specimen C2B** revealed, however, that the fuel

* The post-steady-state-irradiation condition of the fuel was assumed to be quite similar to that of specimen C3A which was steady-state irradiated at about the same conditions as specimen C3B. See Figure 3-5 of the interim report for longitudinal sections of specimen C3A.

** This nonpre-irradiated specimen was subjected to a transient pulse which resulted in approximately the same maximum fuel and cladding temperatures as in specimen C3B. See Figures 3-18 and 3-19 of Reference (4).

FIGURE 3-6. TRANSIENT IRRADIATION DATA, TRANSIENT C3C-1

FIGURE 3-7. CALCULATED RADIAL TEMPERATURE PROFILE IN SPECIMEN C3C FOR MAXIMUM MOLTEN FUEL RADIUS ($t = 2.9$ sec)

TABLE 3-4

DIMENSIONAL MEASUREMENTS SPECIMEN C3B

Location ^(a)	Diameter (inch)			
	Pre-Irradiation ^(b)		Post-Irradiation ^(c)	
	0 Degrees	90 Degrees	0 Degrees	90 Degrees
1.5	0.253	0.253	0.251	0.251
2.5*	0.253	0.253	0.254	0.255
3.5*	0.253	0.253	0.255	0.255
4.5*	0.253	0.253	0.255	0.254
5.5*	0.253	0.253	0.254	0.254
6.5*	0.252	0.253	0.255	0.253
7.5*	0.253	0.252	0.255	0.251
8.5	0.253	0.253	0.253	0.251
9.5	0.253	0.253	0.254	0.251
10.5	0.253	0.253	0.253	0.251
11.5	0.253	0.253	0.253	0.252
12.5	0.253	0.253	0.253	0.251
13.5	0.253	0.253	0.253	0.252
14.5	0.252	0.253	0.252	
15.5	0.252	0.253		

(a) Inches from bottom of fuel pin.

(b) Rounded off to nearest mil.

(c) Accuracy = ± 1 mil.

* Fueled region of pin.

FIGURE 3-8. NEUTRON RADIOGRAPH AND FUEL PIN SCHEMATIC SPECIMEN C3E (63,000 MWd/Te BURNUP)

FIGURE 3-9 NEUTRON RADIOGRAPH AND FUEL PIN SCHEMATIC

(a) C3B GAMMA SCAN PRIOR TO TRANSIENT IRRADIATION

NEG. NO. 32962

(b) C3B GAMMA SCAN FOLLOWING TRANSIENT IRRADIATION

NEG. NO. 37029

FIGURE 3-10. COMPARISON OF C3B GAMMA SCANS BEFORE AND AFTER TRANSIENT IRRADIATION

FIGURE 3-11. PROFILE MEASUREMENTS BEFORE AND AFTER IRRADIATION, SPECIMEN C3B

FIGURE 3-12. SPECIMEN C3B FOLLOWING TRANSIENT IRRADIATION

(b) ALPHA AUTORADIOGRAPH

(REDUCED TO 80 PERCENT FOR REPRODUCTION)

(c) BETA-GAMMA AUTORADIOGRAPH

FIGURE 3-13. LONGITUDINAL SECTION FROM TOP OF SPECIMEN C3B FOLLOWING TRANSIENT IRRADIATION

movement was distinctly different from that observed in the zero burnup case. Specifically, in the zero burnup case (specimen C2B), the top portion of the pin contained a conical central void. In the pre-irradiated case (specimen C3B), the top portion of the pin contains fuel that appears to be essentially 100 percent dense, since the small cavity obviously was caused by shrinkage and downward relocation of the fuel upon cooling. In the bottom portion of the C2B pin, ⁽⁴⁾ the fuel had only partially filled the hole in the hollow UO_2 pellet at the bottom of the fuel column; while in the pre-irradiated pin, the hole in the hollow insulator pellet, the stainless steel spacer, and the cavity in the pressure transducer were filled with fuel. This major difference in fuel redistribution between the zero and low burnup cases suggests that the driving force for movement of molten fuel in the pre-irradiated case is the fission gas that is released during the transient.

A more detailed examination of the longitudinal sections was performed. Shown in Figure 3-13 are the photomacrograph and alpha and beta-gamma autoradiographs of the top longitudinal section from the pin. One of the distinguishing features seen in the photomacrographs is the apparent porosity of the fuel in the center of the column. It is assumed that this is the result of fission gases being trapped in the molten fuel during solidification of the fuel, since steady-state operating conditions precluded fission gas retention in the center portion of the fuel. The alpha autoradiograph illustrates the apparent homogeneity of the mixed oxide; i. e., there does not appear to be any significant segregation of the plutonium. The beta-gamma autoradiograph shown in Figure 3-13(c) illustrates the apparent segregation of fission products as a result of melting of the fuel. This is evidenced by the regions of high beta-gamma activity (light area) adjacent to the cladding as con-

trasted to the relatively low activity (dark area) of the center portions of the fuel. The bright spots dispersed throughout the low activity portions of the fuel are believed due to metallic fission products which have coalesced as a result of melting of the fuel.

Figure 3-14 is a high magnification photomicrograph of the area in the beta gamma autoradiograph where the thickness of the high activity zone (molten boundary limit) decreases. It is interesting to note the change in the molten zone boundary as a function of fuel-cladding gap that this photomicrograph illustrates. From measurements of the photomicrograph it appears that there is about a factor of 6 difference in the fuel-cladding gap width.

Figure 3-15 comprises a photomacrograph and alpha and beta-gamma autoradiographs of a longitudinal section taken from the lower half of the fuel pin. The most salient feature of the photomacrograph [Figure 3-15(a)] is the lack of extensive downward fuel relocation after the fuel had been displaced upward by fission gas pressure. The tapered hollow core of fuel adjacent to the walls of the center void is due to fuel which solidified as it flowed downward. This type of fuel relocation after a significant portion of the fuel was melted, was also observed in the zero burnup specimen C2B. ⁽⁴⁾ It is significant to note, however, that the large mass of once molten fuel above the void did not collapse or fall into the center void.

As in the previous alpha autoradiograph [Figure 3-13(b)], no plutonium segregation is indicated in Figure 3-15(b). The porosity (entrapped fission gases) in the fuel which filled the center void at the top of the section is again discernible, while the remainder of the fuel appears to be relatively dense. The segregation of fission products as a result of the transient fuel melting is

50x, AS-POLISHED

NEG. NO. 9292-04

(REDUCED TO 75% FOR REPRODUCTION)

**FIGURE 3-14. PHOTOMICROGRAPH OF FUEL-CLADDING INTERFACE
IN TOP LONGITUDINAL SECTION, SPECIMEN C3B**

illustrated in the beta-gamma autoradiograph, Figure 3-15(c). The condition of the fuel at the bottom of the fuel column is illustrated in Figure 3-16. The most obvious feature in this photomicrograph is the fact that the hole in the center of the hollow UO_2 insulator pellet has been completely filled with fuel. In view of this fuel relocation, it is apparent that the cladding expansion which occurred was due to fission gas pressure, since there was ample void volume to accommodate the ΔV on melting of the fuel.

As seen in Figures 3-16(b) and (c), the alpha and beta-gamma autoradiographs confirm the presence of fuel in the hollow insulator pellet.

In addition to the longitudinal sections just discussed, two transverse sections were selected for detailed metallographic and ceramographic examinations. The first,

Figure 3-17, is from the bottom of the fuel column and corresponds to the surface at the top of the fuel in Figure 3-16.

Presented in Figure 3-18 are composite diametral photomicrographs of the section in the as-polished (a) and etched (b) conditions. The etchant* attack of the fuel is typical of fuel that has been exposed to NaK or sodium. (12, 13) The most outstanding feature of the composites is the well-defined once-molten region of the fuel whose boundary is characterized by the porosity-free band⁽¹⁴⁾ at about six-tenths of the inner radius of the cladding. Measurements of the molten boundary radius in the photomicrographs and in the beta-gamma autoradiograph [Figure 3-17(c)] show excellent agreement.

* 10 percent HNO_3 , 20 percent H_2O_2 , 70 percent H_2O .

(b) ALPHA AUTORADIOGRAPH
(REDUCED TO 80 PERCENT FOR REPRODUCTION)

(c) BETA-GAMMA AUTORADIOGRAPH

FIGURE 3-15. LONGITUDINAL SECTION FROM LOWER
HALF OF SPECIMEN C3B FOLLOWING
TRANSIENT IRRADIATION

10x AS-POLISHED NEG. NO. 9294-01
(a) PHOTOMACROGRAPH

10x NEG. NO. 9294-02

(b) ALPHA AUTORADIOGRAPH

(REDUCED TO 80 PERCENT FOR REPRODUCTION)

NEG. NO. 9294-03
(c) BETA-GAMMA AUTORADIOGRAPH

FIGURE 3-16. LONGITUDINAL SECTION FROM BOTTOM OF FUEL COLUMN OF SPECIMEN C3B FOLLOWING TRANSIENT IRRADIATION

FIGURE 3-17. TRANSVERSE SECTION FROM BOTTOM OF FUEL COLUMN, SPECIMEN C3B

FIGURE 7.13. INVERSE SECTION FROM BOTTOM OF

FIGURE 3-18. DIAMETRAL COMPOSITE OF TRANSVERSE SECTION FROM BOTTOM OF SPECIMEN C3B FUEL COLUMN

WILLIAM DUNNELL

An illustration of the microstructural changes that can occur in fuel as a result of transient irradiation is afforded by comparison of Figure 3-18 with the composite photomicrograph of specimen C3A* which was steady-state irradiated at about the same conditions as specimen C3B but was not transient irradiated.

In specimen C3B it appears that grain growth has occurred at the outer radius of the fuel, while in specimen C3A the fuel structure has not changed substantially. Note also that the fuel-cladding gap has decreased somewhat in specimen C3B.

Proceeding radially inward, it is seen that the limit of equiaxed grain growth occurs at about the same distance from the cladding inside diameter in each case. In specimen C3A there then follow the long columnar grains which terminate at the center void, while in specimen C3B the columnar grains terminate at the molten boundary. Note also the distribution of fission gas bubbles in the equiaxed and columnar grain regions of the fuel in specimen C3B, and particularly the concentration of bubbles immediately adjacent to the molten boundary as contrasted to the lack of any gas bubble concentrations in specimen C3A. As will be discussed in greater detail further on, one of the assumptions required in the analysis of fission gas behavior during rapid (transient) melting of the fuel is that gas is released only from fuel that becomes molten. The concentration of entrapped gas at the molten boundary radius seen in Figure 3-18 appears to substantiate this assumption.

Figure 3-19 is a photomacrograph and beta-gamma autoradiograph of a transverse section which corresponds to the fuel region between those of Figure 3-13 and Figure 3-15.

*Figure 3-6 of the interim report. (10)

As can be seen, the redistributed fuel which has solidified in the center of the original fuel has formed a separate and distinct structure. This is further illustrated in the photomicrographs shown in Figure 3-20 where it is evident that the fuel in the center has a structure characteristic of once-molten material, and appears to be relatively free of porosity or fission gas bubbles. This again implies release of all the gas in this portion of the fuel. As in Figure 3-10, the molten boundary and the concentration of fission gas bubbles adjacent to it are quite evident. Closer examination of the equiaxed grains and the outer regions of the columnar grains reveal porosity (fission gas bubbles) trails and the image of old grain boundaries. These are further illustrated in the high magnification photomicrograph shown in Figure 3-21. It is interesting to note that, as expected, the length of the porosity trail appears to be extremely sensitive to the temperature, since the trails are much longer in those grains closest to the thermal center of the fuel. From Figure 3-3, it appears that the maximum temperature and temperature gradient in this region of the fuel was about 4800°F (2650°C) and 37,500°F/inch (8210°C/cm), respectively. The maximum amount of time that the fuel was at this condition is estimated to be about 5 and 10 seconds. From the foregoing it is obvious that both grain growth and porosity (fission gas bubbles) migration can, and do, occur in very short times at high temperatures.

Figure 3-22 is composed of high magnification photomicrographs from regions of the fuel as indicated in Figure 3-20. The characteristic structure of once molten fuel is illustrated in Figure 3-22(a), the size and concentration of the fission gas bubbles adjacent to the molten boundary are illustrated in Figure 3-22(b), and the fuel-cladding interface is illustrated in Figure 3-22(c). As can be seen, some fuel-cladding reaction appears to have occurred.

10x AS-POLISHED

NEG. NO. 9373-03

(a) PHOTOMACROGRAPH

10x

NEG. NO. 9373-09

(b) BETA-GAMMA
AUTORADIOGRAPH

**FIGURE 3-19. TRANSVERSE SECTION FROM UPPER PORTION
OF FUEL SPECIMEN C3B**

(a) 100x AS-POLISHED

NEG. NO. 9373-10-15

(REDUCED TO 55 PERCENT FOR REPRODUCTION)

(b) 100x ETCHED

NEG. NO. 9373-42-47

(REDUCED TO 55 PERCENT FOR REPRODUCTION)

FIGURE 3-20. DIAMETRAL COMPOSITE OF TRANSVERSE SECTION FROM TOP PORTION OF SPECIMEN C3B FUEL COLUMN

FIGURE 3-21. HIGH MAGNIFICATION PHOTOMICROGRAPH ILLUSTRATING POROSITY (FISSION GAS BUBBLES) TRAILS AND GRAIN GROWTH

More detailed metallographic examination of the cladding revealed that extensive cracking and grain boundary corrosion had occurred over the fueled length as a result of the deformation during transient irradiation and subsequent entry of NaK into the fuel pin. This is illustrated in Figures 3-23 and 3-24. The lack of ductility at the cladding inner surface after only a short period of exposure was not completely anticipated. Examination of unirradiated cladding (Figure 3-25), cladding from specimen C2B,⁽⁴⁾ which was transient irradiated at about the same conditions as specimen C3B but not steady-state irradiated, and cladding from specimen C3A⁽¹⁰⁾ (Figure 3-26), which was steady-state irradiated at the same conditions as specimen C3B but not transient irradiated, did not reveal the intergranular cracking observed in specimen C3B. The possibility of sensitization of the cladding after steady-state irradiation for only 650 hours at an average temperature of about 700°F seems

unlikely. However, at only 8000 MWd/Te it is also unlikely that the loss of ductility was due to radiation-enhanced damage. Thus, the cracking observed in this specimen remains unexplained in light of current knowledge.

3.2.3 Specimen C3E Post-Irradiation Examination

As mentioned previously, the transient irradiation of specimen C3E resulted in modest conditions of cladding temperature and amount of molten fuel in comparison to specimen C3B. The principal objective of the C3E experiment was to simulate fuel and cladding temperatures postulated to occur during a credible excursion accident in an FCR. Application of the model for fission gas release during a transient, derived from the C3B experiment, predicted that no significant fission gas release and no cladding deformation should occur under

these conditions. Post-transient-irradiation examination of the fuel pin verified these predictions.

Gamma scanning of the capsule following steady-state irradiation (Figure 3-27) and following transient irradiation (Figure 3-28) showed that some fuel movement occurred during the transient. The large activity spike in the fuel region in the post-transient gamma scan suggested the presence of a concentration of metallic fission products.

Capsule disassembly was accomplished in a routine manner. No fission gases were detected in the NaK cover gas as determined by gamma spectrometric analysis for Kr-85. This established that cladding integrity had been maintained during both steady-state and transient irradiation.

The appearance of the fuel following transient irradiation is illustrated in Figure 3-29. Bowing of the radially-unrestrained pin was quite pronounced. However, the neutron radiograph, Figure 3-8, revealed that considerable bowing had occurred during steady-state irradiation.

Dimensional measurements of the fuel pin did not disclose any increase in diameter of the cladding. This was determined by micrometer readings of the as-sectioned pieces since the bow in the pin gave rise to erroneous dial-gage readings.

Sectioning of the pin showed that only a small amount of axial fuel redistribution occurred during the transient. Here again, comparison of the neutron radiograph to the longitudinal sections, Figure 3-29(b), (c), (d), and (e), differentiates fuel movement during the transient from fuel relocation which occurred during steady-state irradiation.

A small amount of fuel on the top of the diaphragm is shown in Figure 3-29(b). The bottom insulator pellet [top of Figure 3-29(b) and bottom of Figure 3-29(c)] is seen to contain fuel which was deposited in the annulus during steady-state irradiation as shown by the neutron radiograph. Fuel movement which occurred during the transient is illustrated in Figures 3-29(d) and (e). It should be noted that axial redistribution of fuel in this manner, which has been observed in previous transient irradiations,⁽⁴⁾ probably results from gravity flow during cooling and solidifying, rather than as a result of fission gases released during the transient.

Alpha and beta-gamma autoradiographs of the fuel sections shown in Figure 3-29 were made. These are illustrated in Figures 3-30 through 3-34. There are several interesting features in these illustrations.

- a. In the beta-gamma autoradiographs, the molten fuel boundary is delineated by the region of relatively low activity (dark area) adjacent to the center void.
- b. In the alpha autoradiographs, there is a distinct enrichment of alpha emitters in the fuel adjacent to the center void and the cladding. Micro-core sampling and chemical analysis (see Appendix C) revealed that the principal reason for the gradients in alpha activity was due to plutonium segregation.
- c. The enrichment of alpha emitters adjacent to the center void appears to coincide with the molten boundary.
- d. The white globule in the center of the fuel in Figure 3-31(a) is agglomerated fission products. Chemical analyses of similar ingots from specimen C3D⁽¹⁰⁾ and rod 12GV of the High Performance UO₂ program⁽¹⁵⁾ have shown that the

principal constituents of the ingots are ruthenium, antimony, palladium, molybdenum, and rhodium.⁽¹⁶⁾ It appears that melting of the fuel is required to cause this massive agglomeration of the metallic fission products.

The large activity spike in the gamma scans, Figure 3-28, coincided with the position of the ingot in Figure 3-31(a).

Detailed ceramographic and metallographic examination of the fuel and cladding was conducted on the longitudinal and transverse sections illustrated in Figures 3-30 through 3-34.

Figure 3-35 is composed of photomicrographs of a transverse section from the longitudinal section shown in Figure 3-30(a). The most significant point of interest in Figure 3-35 is the distinct molten boundary which corresponds to the boundary delineated in Figure 3-30. It is obvious from Figure 3-35 that this molten boundary corresponds to the outside edge of the high density pore-free region. Further, it is seen that the dispersion of metallic fission products terminates at the molten boundary. This is in agreement with and lends additional support to Lyons' definition of the molten boundary in UO_2 .⁽¹⁴⁾

Several other interesting points to be noted in Figure 3-35 are:

- a. The outer radius of the fuel is extremely porous, which is typical of highly irradiated oxide fuel.⁽¹³⁾
- b. The fuel-cladding gap has been closed. It is believed this occurred during steady-state irradiation and is at least partially a result of fission product swelling since previous transient irradiations of nonpre-irradiated mixed oxide fuel under

more severe conditions (in terms of volume of molten fuel) did not result in closing of the fuel-cladding gap.⁽⁴⁾

- c. Some evidence of fuel-cladding interface reaction is present.

Figure 3-36 is a diametral photomicrograph composite of the longitudinal section shown in Figure 3-31. Except for the outer regions of the fuel being attacked by the etchant, the microstructural details are very similar to those in Figure 3-35.

Illustrated in Figure 3-37 is the microstructure of the relocated fuel shown in the photomicrograph in Figure 3-32. Note the unusual porosity in the once-molten fuel and the definition of the molten boundary.

Figures 3-38 and 3-39 are composite diametral photomicrographs of the transverse section illustrated in Figure 3-33. Inspection of these illustrations shows that:

- a. The molten boundary is easily discernible even in the as-polished condition since it can be defined as the termination of the dispersion of the metallic fission products.
- b. It appears that the most pronounced concentration of metallic fission products occurs in the columnar grain region of the fuel. This would seem to indicate a strong temperature dependence for the migration and coalescence of these fission products.
- c. The outer region of the fuel is extremely porous and quite susceptible to attack by the etchant.
- d. An amorphous grey phase, seen in the fuel-cladding interface in the as-polished condition [Figure 3-38(d)], seems to be relatively unaffected by the

etchant as it is still present in Figure 3-39(a).

- e. Some fuel-cladding reaction has occurred, but this appears to be only of superficial extent.

Figures 3-40 and 3-41 are composite diametral photomicrographs of the transverse section illustrated in Figure 3-34. As can be seen, the details in these illustrations are quite similar to those of previous sets. The rather massive grey phase seen in the area adjacent to the cladding appears to be quite resistant to the etchant. An electron microprobe study of such phases in mixed oxide fuel is being conducted by another task in the FCR development program.

Metallographic examination of the cladding, Figure 3-42, showed nonuniform grain boundary precipitation on the inside diameter of the cladding. The reason for the nonuniformity is not known, but it may be related to differences in the cladding temperatures (and stress levels) which could have been brought about by the bowing of the fuel pin. This circumferential variation in cladding microstructure occurred over the entire length of the fueled portion of the pin. Further study of the grain boundary precipitation, the fission-product recoil layer, and the fuel-cladding interaction zones by means of electron microscopy and electron microprobe is being conducted by another task in the FCR development program.

Collection and analysis of the fission gases in the fuel pin were performed. The results obtained from gamma spectrometry and gas chromatography analyses are presented in Table 3-5.

TABLE 3-5

GAS COMPOSITION AND VOLUME
IN SPECIMEN C3E

<u>Gas</u>	<u>Volume Percent</u>
Krypton	5.0 ± 0.3*
Xenon	84.4 ± 4.2*
Hydrogen	0.1
Nitrogen	0.1
Oxygen and/or Argon	0.3
Helium	Balance
Kr-85	1.1×10^{17} atoms/cm ³

Qualitative
Estimates

Total gas in the fuel pin (at STP) was determined to be 69.2 cm³.

*Limits represent estimated precision.

The isotopic analysis of the fission gas is presented in Table 3-6.

TABLE 3-6

ISOTOPIC ANALYSIS OF FISSION GAS
IN SPECIMEN C3E

<u>Gas</u>	<u>Atom Percent</u>
Kr-83	19.8 ± 4.0
Kr-84	27.7 ± 5.5
Kr-85	9.0 ± 1.8
Kr-86	43.5 ± 8.7
Xe-131	14.5 ± 2.9
Xe-132	20.5 ± 4.1
Xe-134	25.7 ± 5.1
Xe-136	39.3 ± 7.9

Radiochemical and mass spectrometer analyses for fuel burnup were conducted. These data are presented in Table 3-7. The

500x ETCHED

NEG. NO. 9373-50

(a) FUEL AT EDGE OF CENTER VOID

500x ETCHED

NEG. NO. 9373-49

(b) FUEL IN AREA OF CONCENTRATED POROSITY
(FISSION GAS BUBBLES)

← THERMAL CENTER

CLADDING →

500x ETCHED

NEG. NO. 9373-48

(c) FUEL-CLADDING INTERFACE

FIGURE 3-22. HIGH MAGNIFICATION PHOTOMICROGRAPHS FROM REGIONS OF FUEL INDICATED IN FIGURE 3-18

NEG. NO. 9294-04

(a) 100x AS-POLISHED

NEG. NO. 9294-05

(b) 500x AS-POLISHED

(REDUCED TO 85 PERCENT FOR REPRODUCTION)

NEG. NO. 9294-06

(c) 100x 10% OXALIC ACID ETCH

NEG. NO. 9294-07

(d) 500x 10% OXALIC ACID ETCH

FIGURE 3-23. PHOTOMICROGRAPHS ILLUSTRATING CRACKING AND GRAIN BOUNDARY CORROSION IN CLADDING FROM BOTTOM LONGITUDINAL SECTION OF SPECIMEN C3B

(a) FUEL CLADDING REACTION IN TRANSVERSE CROSS SECTION ILLUSTRATED IN FIGURE 3-18

(b) CLADDING CRACKING AT CLADDING INSIDE DIAMETER IN TRANSVERSE CROSS SECTION ILLUSTRATED IN FIGURE 3-18

(c) CRACKS IN CLADDING IN TRANSVERSE CROSS SECTION ILLUSTRATED IN FIGURE 3-20

(REDUCED TO 85 PERCENT FOR REPRODUCTION)

(d) SAME AS (c) EXCEPT ETCHED AND HIGHER MAGNIFICATION

FIGURE 3-24. PHOTOMICROGRAPHS ILLUSTRATING FUEL-CLADDING REACTIONS AND CLADDING CRACKING IN SPECIMEN C3B

NEG. NO. 65C2310

(a) LONGITUDINAL SECTION OF CLADDING

500x
10% OXALIC
ACID ETCH

NEG. NO. 65C2311

(b) HIGH MAGNIFICATION PHOTOMICROGRAPH
OF UNIRRADIATED TYPE 347 STAINLESS
STEEL CLADDING

**FIGURE 3-25. METALLOGRAPHIC EXAMINATION OF UNIRRADIATED
TYPE 347 STAINLESS STEEL CLADDING**

**FIGURE 3-26. SPECIMEN C3A CLADDING STRUCTURE
FOLLOWING STEADY-STATE IRRADIATION**

FIGURE 3-27. POST-STEADY-STATE-IRRADIATION
GAMMA SCAN OF CAPSULE C3E

427

(a)

(b)

(c)

FIGURE 3-28. POST-TRANSIENT IRRADIATION GAMMA SCANS OF CAPSULE C3E

FIGURE 3-29. SPECIMEN C3E FOLLOWING STEADY-STATE AND TRANSIENT IRRADIATION

NEG. NO. 9370-02

(a) PHOTOMACROGRAPH
AS-POLISHED 10x

NEG. NO. 9370-04

(b) BETA-GAMMA AUTORADIOGRAPH

(REDUCED TO 80 PERCENT FOR REPRODUCTION)

NEG. NO. 9370-03

(c) ALPHA AUTORADIOGRAPH

FIGURE 3-30. PHOTOMACROGRAPH AND BETA-GAMMA
AND ALPHA AUTORADIOGRAPHS OF
LONGITUDINAL SECTION FROM BOTTOM
OF SPECIMEN C3E

NEG. NO. 9371-01

(a) PHOTOMACROGRAPH
AS-POLISHED 10x

NEG. NO. 9371-03

(b) BETA-GAMMA AUTORADIOGRAPH

(REDUCED TO 80 PERCENT FOR REPRODUCTION)

NEG. NO. 9371-02

(c) ALPHA AUTORADIOGRAPH

FIGURE 3-31. PHOTOMACROGRAPH AND BETA-GAMMA
AND ALPHA AUTORADIOGRAPHS OF
LONGITUDINAL SECTION FROM CENTER
OF SPECIMEN C3E

AS-POLISHED 100x NEG. NO. 9372-01
(a) PHOTOMACROGRAPH

NEG. NO. 9372-03

(b) BETA-GAMMA AUTORADIOGRAPH

NEG. NO. 9372-02

(c) ALPHA AUTORADIOGRAPH

(REDUCED TO 80 PERCENT FOR REPRODUCTION)

FIGURE 3-32. PHOTOMACROGRAPH AND BETA-GAMMA AND ALPHA AUTORADIOGRAPHS OF LONGITUDINAL SECTION FROM TOP OF SPECIMEN C3E

NEG. NO. 9373-01

(a) PHOTOMACROGRAPH
10x AS-POLISHED

NEG. NO. 9373-08

(b) BETA-GAMMA AUTORADIOGRAPH

NEG. NO. 9373-04

(c) ALPHA AUTORADIOGRAPH

(REDUCED TO 75% FOR REPRODUCTION)

**FIGURE 3-33. PHOTOMACROGRAPH AND BETA-GAMMA AND ALPHA AUTORADIOGRAPHS OF
TRANSVERSE SECTION FROM CENTER OF SPECIMEN C3E**

NEG. NO. 9373-02

(a) PHOTOMACROGRAPH
10x AS-POLISHED

NEG. NO. 9373-07

(b) BETA-GAMMA AUTORADIOGRAPH

NEG. NO. 9373-05

(c) ALPHA AUTORADIOGRAPH

GEAP-4469

(REDUCED TO 75 PERCENT FOR REPRODUCTION)

FIGURE 3-34. PHOTOMACROGRAPH AND BETA-GAMMA AND ALPHA AUTORADIOGRAPHS OF
TRANSVERSE SECTION FROM TOP PORTION OF SPECIMEN C3E

(a) COMPOSITE PHOTOMICROGRAPH OF TRANSVERSE SECTION FROM THE LONGITUDINAL SECTION SHOWN IN FIGURE 3-28

(b) PHOTOMICROGRAPH OF AREA INDICATED IN (a)

(REDUCED TO 55 PERCENT FOR REPRODUCTION)

FIGURE 3-35. PHOTOMICROGRAPH ILLUSTRATING MOLTEN BOUNDARY RADIUS AND DISTRIBUTION OF METALLIC FISSION PRODUCTS IN LONGITUDINAL SECTION SHOWN IN FIGURE 3-30

0.015 in.
CLADDING

(a) COMPOSITE PHOTOMICROGRAPH OF TRANSVERSE SECTION FROM LONGITUDINAL SECTION SHOWN IN FIGURE 3-31

ETCHED 100X

NEG. NO. 9371-11-16

(REDUCED TO 60 PERCENT FOR REPRODUCTION)

ETCHED 500x

NEG. NO. 9371-17

(b) PHOTOMICROGRAPH OF AREA INDICATED IN (a)

FIGURE 3-36. PHOTOMICROGRAPHS ILLUSTRATING MOLTEN BOUNDARY RADIUS AND DISTRIBUTION OF METALLIC FISSION PRODUCTS IN LONGITUDINAL SECTION SHOWN IN FIGURE 3-31

NEG. NO. 9372-13

(a) 100x PHOTOMICROGRAPH OF AREA INDICATED
IN FIGURE 3-32

NEG. NO. 9372-14

(b) HIGH MAGNIFICATION PHOTOMICROGRAPH OF
AREA SHOWN IN (a)

**FIGURE 3-37. PHOTOMICROGRAPHS OF AREA OF RELOCATED FUEL
SHOWN IN FIGURE 3-32 (SPECIMEN C3E)**

FIGURE 3-38. COMPOSITE PHOTOMICROGRAPH AND HIGH MAGNIFICATION PHOTOMICROGRAPHS OF TRANSVERSE CROSS SECTION SHOWN IN FIGURE 3-33 (SPECIMEN C3E, AS-POLISHED)

(a) COMPOSITE DIAMETRAL PHOTOMICROGRAPH 100x ETCHED

NEG. NO. 9373-53-58

500x ETCHED

NEG. NO. 9373-59

(b) FUEL-CLADDING INTERFACE

500x ETCHED

NEG. NO. 9373-60

(c) FUEL AT CENTER

500x ETCHED

NEG. NO. 9373-61

(d) FUEL AT $\frac{3}{4}$ RADIUS

(REDUCED TO 55 PERCENT FOR REPRODUCTION)

FIGURE 3-39. COMPOSITE PHOTOMICROGRAPH AND HIGH MAGNIFICATION PHOTOMICROGRAPHS OF TRANSVERSE CROSS SECTION SHOWN IN FIGURE 3-33 (SPECIMEN C3E, ETCHED)

(REDUCED TO 55 PERCENT FOR REPRODUCTION)

(a) COMPOSITE DIAMETRAL
PHOTOMICROGRAPH
100X

(b) FUEL AT EDGE
OF CENTER VOID
500X

(d) GREY PHASE
500X

(e) FUEL-CLADDING
INTERFACE
500X

FIGURE 3-40. COMPOSITE PHOTOMICROGRAPH AND HIGH
MAGNIFICATION PHOTOMICROGRAPHS OF
TRANSVERSE CROSS-SECTION SHOWN IN
FIGURE 3-34 (SPECIMEN C3E, AS-POLISHED)

FIGURE 3-41. COMPOSITE PHOTOMICROGRAPH AND HIGH MAGNIFICATION PHOTOMICROGRAPHS OF TRANSVERSE CROSS-SECTION SHOWN IN FIGURE 3-34 (SPECIMEN C3E, ETCHED)

(a) PHOTOMICROGRAPH SHOWING GRAIN
BOUNDARY PRECIPITATE ON ID
(250X ETCHED)

(b) PHOTOMICROGRAPH SHOWING NO GRAIN
BOUNDARY PRECIPITATE ON ID
(250X ETCHED)

**FIGURE 3-42. METALLOGRAPHIC EXAMINATION OF
SPECIMEN C3E CLADDING (347 SS)**

Cs-137/He ratio is low, due probably to the redistribution of the cesium during steady-state irradiation.⁽¹⁷⁾ On the other hand, the mass spectrometric data appear to agree quite well with the thermocouple data given in Table 3-1.

TABLE 3-7
SPECIMEN C3E BURNUP DATA

Ratio	Burnup (percent)	MWd/Te*
Ce-144/He	7.14	66,700
Cs-137/He	4.90	45,700
<u>Mass Spectrometer</u>		
Pu-239	6.91	
Pu-241	0.17	
U-235	0.10	
Total	7.18	67,000

*Based on 200 MeV/fission.

Estimates of the total fission gas generated when compared to the measured quantity of fission gases collected indicate that the gas release was in the range of 80 to 93 percent. Detailed calculations for the estimation of fission gas release are included in Appendix C.

3.2.4 Specimen C3C Post-Irradiation Examination

As discussed in subsection 3.1.3, the objectives of the transient irradiation of this second high burnup specimen were twofold: a.) to establish the transient performance limit for high burnup fuel, and b.) to verify the fission gas release model deduced from the C3B experiment and demonstrate the lack of cladding ductility which was also first observed in the C3B experiment. Design of the transient pulse was based on melting a sufficient quantity of fuel to release enough fission gas to cause approximately 1 percent cladding deformation.

Prior to re-encapsulating the specimen for transient irradiation, an attempt was made to sample the steady-state released fission gas in the pin by puncturing the fuel pin diaphragm* with a bellows and hypodermic needle connected to a fission gas sampling system. Unfortunately the attempt was not successful as no fission gases were collected, even after the bellows had been fully collapsed. Thus, either the diaphragm was punctured and the hypodermic needle was plugged, the diaphragm was not punctured, or the fission gases escaped without detection. The latter possibility is remote, since RML off-gas monitors gave no indication of increased activity. Since the neutron radiograph, Figure 3-9, showed that the hollow insulator pellet contained fuel which had relocated during steady-state irradiation, it is possible that plugging of the needle occurred.

Transient irradiation of the specimen was accomplished without incident, but comparison of the pre- and post-transient gamma scans, Figure 3-43, revealed that extensive downward fuel relocation had occurred during the transient.

When the bottom section of the capsule was punctured, fission gases were detected, indicating fuel pin failure had occurred. Subsequent disassembly of the capsule revealed the nature and extent of the failure seen in Figure 3-44. Molten fuel was ejected through the hypodermic needle into the bellows assembly where melting of the needle and the bellows permitted expulsion of the fuel into the void between the check valve and the housing. Approximately 11 grams of fuel were recovered from the housing. The total fuel inventory was about 38 grams. It was concluded qualitatively, that molten fuel is apparently extremely fluid, inasmuch as the hypodermic needle internal diameter was only 0.033 inch.

* See Figure 3-44 for construction details.

a) C3C GAMMA SCAN FOLLOWING STEADY-STATE IRRADIATION TO 64,000 MWD/MT BURNUP

b) C3C GAMMA SCAN FOLLOWING TRANSIENT IRRADIATION

FIGURE 3-43. GAMMA SCANS OF CAPSULE C3C

FIGURE 3-44. FUEL REDISTRIBUTION FOLLOWING TRANSIENT IRRADIATION OF SPECIMEN C3C

It is shown in Figure 3-45 that the external condition of the fuel pin was relatively unaffected by the long steady-state irradiation and subsequent transient irradiation. The as-cut sections, however, revealed evidence of the molten fuel having been expelled from the pin, leaving a very large central void and a thin shell of fuel which adhered to the cladding.

Dimensional measurements of the pin prior to sectioning revealed that expansion of the cladding over the fueled length had occurred. As seen by the data presented in Table 3-8, the average diametral expansion of the cladding over the fueled length was approximately 2.3 mils or about 0.9 percent. This compared favorably with the target value of 1 percent deformation.

Metallographic and ceramographic examination of the cladding and fuel, respectively, was conducted. Photomacrographs

of the longitudinal and transverse sections taken from the positions indicated on the schematic sketch are presented in Figure 3-46. The extent of the fuel expulsion through the bottom of the pin is again illustrated in the longitudinal sections. Since the expansion of the cladding is attributed to the fission gas pressure resulting from release of gas during the transient, it follows that the principal driving force for expulsion of the fuel was also the fission gas pressure.

Presented in Figure 3-47 are the alpha and beta-gamma autoradiographs of the longitudinal section shown in Figure 3-46(c). The molten boundary diameter as measured from the beta-gamma autoradiograph was 0.180 inch. The unusual fuel structure could be a result of fuel flow while cooling, after the initial expulsion under fission gas pressure.

TABLE 3-8
SPECIMEN C3C DIAMETER MEASUREMENTS

Reference Point	0 Degrees		90 Degrees	
	Pre-Irradiation	Post-Irradiation	Pre-Irradiation	Post-Irradiation
1	0.2526	0.2525	0.2527	0.2525
2*	0.2526	0.2550	0.2525	0.2550
3*	0.2526	0.2555	0.2525	0.2550
4*	0.2526	0.2550	0.2524	0.2550
5*	0.2525	0.2550	0.2523	0.2545
6*	0.2526	0.2550	0.2524	0.2545
7*	0.2525	0.2550	0.2524	0.2545
8*	0.2525	0.2545	0.2523	0.2535
9	0.2526	0.2525	0.2525	0.2530
10	0.2527	0.2525	0.2525	0.2525
11	0.2528	0.2525	0.2526	0.2525
12	0.2527	0.2525	0.2527	0.2525
13	0.2527	0.2525	0.2526	0.2525
14	0.2524	0.2525	0.2523	0.2525
15	0.2530	0.2525	0.2529	0.2525

*Fueled length.

FIGURE 3-45. FUEL PIN EXTERIOR AND AS-CUT SECTIONS FROM SPECIMEN C3C

FIGURE 3-46. LONGITUDINAL AND TRANSVERSE SECTIONS FROM SPECIMEN C3C

NEG. NO. 9461-01
(a) PHOTOMACROGRAPH

NEG. NO. 9461-02
(b) ALPHA AUTORADIOGRAPH
(REDUCED TO 80 PERCENT FOR REPRODUCTION)

NEG. NO. 9461-03
(c) BETA-GAMMA AUTORADIOGRAPH

FIGURE 3-47. PHOTOMACROGRAPH AND ALPHA AND BETA-GAMMA
AUTORADIOGRAPHS FROM LONGITUDINAL SECTION
FROM SPECIMEN C3C

100X AS-POLISHED

(a) FUEL AND CLADDING PHOTOMICROGRAPH

NEG. No. 9461-10

250X ETCHED

(b) CLADDING PHOTOMICROGRAPH

NEG. No. 9461-08, 09

FIGURE 3-48. PHOTOMICROGRAPHS OF FUEL AND CLADDING FROM LONGITUDINAL SECTION OF SPECIMEN C3C SHOWN IN FIGURE 3-47.

Figure 3-48 is a high magnification photomicrograph of the fuel and cladding from a section of Figure 3-47. In Figure 3-48(a), the fuel microstructure shows the characteristic porosity in the outer region and the termination of the porosity (molten boundary) in the inner region. Some evidence of fuel-cladding reaction is also discernible. The grain boundary precipitation in the cladding inside surface is shown in Figure 3-48(b).

Illustrated in Figures 3-49, 3-50, and 3-51 is the condition of the fuel and cladding in the transverse sections shown in Figure 3-46(e), (f), and (g). The salient features are:

- a. The molten fuel boundary as delineated by the alpha and beta-gamma autoradiographs and the fuel microstructure.
- b. The grain boundary precipitation at the cladding inside surface and the cracking which appears to be initiated at the grain boundaries.
- c. In the as-polished photomicrographs of the fuel-cladding interface, it is interesting to note that the cracks in the

cladding are associated with cracks in the fuel. This could possibly be explained by assuming that the fuel is in such contact with the cladding so as to prohibit relative tangential motion of the fuel and cladding. When the pin is subjected to a transient, the internal pressure resulting from fission gases released during the transients causes cladding deformation to occur. Since the fuel is well into the plastic temperature range at this time, it flows and assumes the shape of the cladding. As cooling occurs, the relative tangential motion between fuel and cladding, required to accommodate the different expansion characteristics of the fuel and cladding, is restrained by the fuel-cladding interaction forces. Thus, the fuel must crack radially to relieve the shrinkage stresses, and cracking will occur at the point where the fuel-cladding interface forces are a minimum, namely at the cracks in the cladding.

Attempts to collect the fission gases in the bottom portion of the capsule after the transient irradiation were not successful. Thus, no fission gas data are available from this experiment.

NEG. NO. 9459-01

(a) PHOTOMACROGRAPH
10X AS-POLISHED

NEG. NO. 9459-04

(b) ALPHA AUTORADIOGRAPH 10X

NEG. NO. 9459-07

(c) BETA-GAMMA AUTORADIOGRAPH 10X

NEG. NO. 9459-19

(d) PHOTOMICROGRAPH OF FUEL-CLADDING 250X AS-POLISHED

NEG. NO. 9459-21

(e) PHOTOMICROGRAPH OF CLADDING INSIDE DIAMETER 250X ETCHED

FIGURE 3-49. PHOTOMACROGRAPHS, PHOTOMICROGRAPHS, AND ALPHA AND BETA-GAMMA AUTORADIOGRAPHS FROM TRANSVERSE SECTION OF SPECIMEN C3C SHOWN IN FIGURE 3-46e

10X AS-POLISHED
(a) PHOTOMACROGRAPH
NEG. NO. 9459-02

10X
(b) ALPHA AUTORADIOGRAPH
NEG. NO. 9459-05

10X
(c) BETA-GAMMA AUTORADIOGRAPH
NEG. NO. 9459-08

250X AS-POLISHED
(d) FUEL-CLADDING INTERFACE
NEG. NO. 9459-20

FUEL

CLADDING

i.d. CLADDING

250X ETCHED
(e) CLADDING INSIDE DIAMETER
NEG. NO. 9459-23

FIGURE 3-50. PHOTOMACROGRAPH, PHOTOMICROGRAPHS, AND ALPHA AND BETA-GAMMA AUTORADIOGRAPHS OF TRANSVERSE SECTION FROM SPECIMEN C3C SHOWN IN FIGURE 3-46f

10X AS-POLISHED| NEG. NO. 9459-03
(a) PHOTOMACROGRAPH

NEG. NO. 9459-06
(b) ALPHA AUTORADIOGRAPH

NEG. NO. 9459-09
(c) BETA-GAMMA AUTORADIOGRAPH

100X AS-POLISHED| NEG. NO. 9459-15
(d) FUEL-CLADDING INTERFACE

250X AS-POLISHED| NEG. NO. 9459-16
(e) CRACKS IN CLADDING AND FUEL-CLADDING INTERFACE

250X ETCHED| NEG. NO. 9459-24
(f) CRACKS IN CLADDING AND GRAIN BOUNDARY PRECIPITATE

FIGURE 3-51. PHOTOMACROGRAPH, PHOTOMICROGRAPHS, AND ALPHA AND BETA-GAMMA AUTORADIOGRAPHS FROM TRANSVERSE SECTION FROM SPECIMEN C3C SHOWN IN FIGURE 3-46g

SECTION IV

DISCUSSION OF RESULTS

4.1 DERIVATION OF TRANSIENT FISSION GAS RELEASE MODEL FROM THE C3B EXPERIMENT RESULTS

The unique results obtained in the C3B experiment, viz., cladding expansion only over the fueled length and axial fuel relocation both upward and downward, suggested that fission gases released from the fuel during the transient were not immediately vented to the plenum. The following analysis was conducted to correlate measured cladding deformation with the computed internal pressure resulting from the release of fission gases during the transient.

4.1.1 Steady-State Fission Gas Release

The first step in determining the internal pressure was to estimate the steady-state gas release. From physics calculations the power generation profile in the fuel was determined as shown in Figure 4-1. The equation of this curve, in terms of a sixth-power polynomial, was determined from a least squares computer routine.

If it is assumed that the fission gas is generated in direct proportion to the power profile, then a gas generation profile can be obtained from the expression

$$\text{Percent gas} = \frac{\int_a^r r f(r) dr}{\int_a^b r f(r) dr} \quad (100),$$

where

$$f(r) = 0.25775 - 0.535588 r + 128.811 r^2 - 4,662.41 r^3 + 88,926 r^4 - 738,285 r^5 + 2,355,430 r^6,$$

a = Inner radius of fuel pellet, and
b = Outer radius of fuel pellet.

The results of this calculation are presented in Figure 4-2.

It is known that fission gas release in ceramic fuel is a function of several parameters including temperature. A number of workers^(18, 19, 20) have proposed various percentages of gas release for various temperature zones in the fuel. The initial model used in this analysis is that cited by O'Neill,⁽²⁰⁾ where gas release is assumed to be related to fuel temperature by:

$$\begin{aligned} &100 \text{ percent release for} \\ &\quad T \geq 1800^\circ\text{C} \quad (3272^\circ\text{F}) \\ &50 \text{ percent release for} \\ &\quad 1600^\circ\text{C} \leq T \leq 1800^\circ\text{C}, \text{ and} \\ &4 \text{ percent release for} \\ &\quad T \leq 1600^\circ\text{C} \quad (2912^\circ\text{F}). \end{aligned}$$

To verify the validity of the model, the calculated gas release was compared to the measured gas release (15 percent) from Specimen C3A.⁽¹⁰⁾ The calculations (which are presented in Appendix B) showed that this model over-predicted the steady-state gas release. Accordingly, the model was arbitrarily adjusted so that gas release was related to fuel temperature by:

$$\begin{aligned} &100 \text{ percent release for} \\ &\quad T \geq 2400^\circ\text{C} \quad (4350^\circ\text{F}), \\ &50 \text{ percent release for} \\ &\quad 2200^\circ\text{C} \leq T \leq 2400^\circ\text{C}, \text{ and} \\ &4 \text{ percent release for} \\ &\quad T \leq 2200^\circ\text{C} \quad (4000^\circ\text{F}). \end{aligned}$$

Determination of the steady-state radial temperature profile in the fuel involves calculating $\int_{T_s}^{T_r} k dT$ for a fuel pellet with a severe radial power depression. This was accomplished using the expression derived in Appendix C. The linear heat generation rate was taken as

FIGURE 4-1. CALCULATED POWER PROFILE IN SPECIMEN C3B

FIGURE 4-2. FISSION GAS GENERATION IN SPECIMEN C3B

18.1 kW/ft (Table 3-1). From the heat transfer calculations given in the interim report (pp B-1 to B-6), the fuel surface temperature was determined by

$$\begin{aligned} T_s &= \Sigma \Delta T + T_w \\ &= \frac{18.1}{29.3} (2210) + 110 \\ &= 1475^\circ\text{F} \quad (801^\circ\text{C}), \end{aligned}$$

where

$\Sigma \Delta T$ = The summation of the temperature drops in the capsule from the coolant to the fuel surface, and

T_w = The coolant temperature.

From Figure 4-3, which is an $\int k dT$ curve for mixed oxide fuel,⁽²¹⁾ and the computer calculations for $\int_{T_s}^T r k dT$, the radial temperature profile shown in Figure 4-4 was established. By superimposing the temperature profile plot for fission gas generation, the fraction of fission gas released was determined. From Figure 4-5, it is seen that no fuel experienced a temperature greater than 4350°F. Thus, it was assumed that none of the fuel released 100 percent of the fission gas. At $T = 4000^\circ\text{F}$, $r = 0.041$ inch, which corresponds to 5.4 percent of the gas content. The steady-state gas release was:

$$\begin{aligned} \text{From the 50 percent zone:} \\ (0.5)(5.4) &= 2.7 \text{ percent} \\ \text{From the 4 percent zone:} \\ (0.04)(100-5.4) &= 3.8 \text{ percent} \\ \text{Total steady-state release} &= 6.5 \text{ percent} \end{aligned}$$

4.1.2 Fission Gas Released During Transient

For the purpose of this analysis in view of the fact that workers in the field^(18, 19, 20, 22, 23) have shown that fission gas release from ceramic fuel is a very complex time-temperature dependent phenomenon, it was assumed that during a transient, fission gas would be released only from fuel that becomes molten. From the post-irradiation examination of the fuel, it was determined that the molten boundary radius was 0.072 inch.* From Figure 4-5, this molten boundary extended into the zone of the fuel that had released only 4 percent of the gas generated during the steady-state irradiation. The gas released during the transient was:

$$\begin{aligned} \text{From the 50 percent zone:} &= 2.7 \text{ percent} \\ \text{From the 4 percent zone:} \\ (0.96)(26-5.4) &= 19.8 \text{ percent} \\ \text{Total transient release} &= 22.5 \text{ percent} \end{aligned}$$

4.1.3 Estimation of Internal Pressure in Fuel Pin Due to Fission Gas Release During Transient

Based on fuel porosity, fuel-cladding gap, and gas plenum, a total internal void volume of 0.232 in.³ was calculated for the pin. The installation of the pressure transducer after the steady-state irradiation added 0.073 in.³, so that the total void volume prior to transient irradiation was 0.305 in.³.

From Table 3-1, the burnup for specimen C3B was determined to be 8100 MWd/Te. From the derivation given in Appendix C, 31.3 percent of the fissions result in the formation of fission gases. Therefore, the amount of fission gas formed during steady-state irradiation can be found as follows:

*See Figures 3-17 and 3-18.

FIGURE 4-3. Kd AND K VERSUS TEMPERATURE

FIGURE 4-4. CALCULATED STEADY-STATE TEMPERATURE PROFILE IN SPECIMEN C3B, $Q' = 18.1$ kW/ft

FIGURE 4-5. STEADY-STATE TEMPERATURE PROFILE AND FISSION GAS GENERATION, SPECIMEN C3B

- a. Number of fissions during steady-state irradiation.

$$N_f = \left[\text{B. U. (MWd/Te)} \right] \left[\frac{\text{Number of Fissions}}{\text{MWd}} \right] \left[10^{-6} \text{ Te/gm} \right] \left[\text{Weight of fuel (gm)} \right] \left[\frac{\text{Fraction of fuel which is fissionable (U-235 and Pu-239)}}{\text{fissionable (U-235 and Pu-239)}} \right]$$

$$N_f = \left[8100 \text{ MWd/Te} \right] \left[2.696 \times 10^{21} \text{ fissions/MWd} \right] \left[10^{-6} \text{ Te/gm} \right] \left[38.84 \text{ gm PuO}_2\text{-UO}_2 \right] \left[0.881 \frac{\text{gm Pu-U}}{\text{gm PuO}_2\text{-UO}_2} \right],$$

$$N_f = 7.46 \times 10^{20} \text{ fissions during steady-state irradiation.}$$

- b. Number of fission gas atoms formed.

$$N_{fg} = \left[0.313 \frac{\text{fission gas atoms}}{\text{fission}} \right] \left[7.46 \times 10^{20} \text{ fissions} \right].$$

$$N_{fg} = 2.34 \times 10^{20} \text{ fission gas atoms.}$$

- c. Volume of fission gas formed at STP.

$$V_{fg} = \left[2.34 \times 10^{20} \text{ fission gas atoms} \right] \left[\frac{2.24 \times 10^4 \text{ cm}^3/\text{gm-mole}}{6.023 \times 10^{23} \frac{\text{atoms}}{\text{gm-mole}}} \right]$$

$$V_{fg} = 8.71 \text{ cm}^3 \text{ at STP.}$$

From subsections 4.1.1 and 4.1.2, we obtain the fractions of steady-state and transient fission gas release.

$$\begin{aligned} \text{Steady-state fission gas release} \\ &= (6.5 \text{ percent})(8.71 \text{ cm}^3) \\ &= 0.566 \text{ cm}^3 \text{ at STP.} \end{aligned}$$

$$\begin{aligned} \text{Transient fission gas release} \\ &= (22.5 \text{ percent})(8.71 \text{ cm}^3) \\ &= 1.96 \text{ cm}^3 \text{ at STP.} \end{aligned}$$

At the completion of the fabrication, the fuel pin was filled with helium (0.232 in.^3) at 1 atmosphere pressure. Thus, the room temperature pressure after steady-state irradiation was:

$$P = 1.0 + \left[\frac{0.566 \text{ cm}^3}{(0.232 \text{ in.}^3)(16.39 \text{ cm}^3/\text{in.}^3)} \right] \left[\frac{520^\circ\text{R}}{492^\circ\text{R}} \right],$$

$$P = 1.16 \text{ atm.}$$

Addition of the pressure transducer volume, originally at a pressure of 1 atmosphere of helium, decreased this to

$$(1.16) \left(\frac{0.232}{0.305} \right) + \left(\frac{0.073}{0.305} \right) (1.0) = 1.06 \text{ atm.}$$

Prior to the initiation of the transient pulse in TREAT, the capsule was heated to 800°F (1260°R). The pressure in the fuel pin at this temperature was

$$(1.06) \left(\frac{1260}{520} \right) = 2.72 \text{ atm.}$$

Now, if it is assumed, on the basis of the post-irradiation examination results, that the fission gases released from the fuel during the transient are not vented to the gas plenum but are retained in the fueled length of the pin, then there are 1.96 cm^3 of gas (at STP) that are released to the void volume in the fueled length. This void volume was calculated to be 0.28 cm^3 , and is based on the assumption that all the void in the fuel and between the fuel

and cladding is available to accommodate the fission gases. It also implies that the fuel is plastic enough to yield rapidly and thus give up the occluded porosity. A further assumption required is that the released gas is at the temperature of the molten fuel. At a temperature of 800°F (1260°R) and pressure of 2.72 atmospheres, the 1.96 cm³ of gas at STP becomes

$$(1.96) \left(\frac{1}{2.72} \right) \left(\frac{1260}{492} \right) = 1.85 \text{ cm}^3.$$

Raising the temperature of the gas to 4900°F (5360°R) compressing it to 0.28 cm³ (available volume), and adding the contribution from the original gas in the fuel section, results in an internal pressure of

$$\begin{aligned} P &= (2.72) \left[\frac{5360}{1260} \right] \left[\left(\frac{1.85}{0.28} \right) + 1 \right] = 88.0 \text{ atm,} \\ &= 1292 \text{ psia,} \\ &= 1277 \text{ psig.} \end{aligned}$$

4.1.4 Estimation of Internal Pressure in Fuel Pin from Cladding Deformation

As seen in Table 3-4, the measured deformation of the cladding was 3 mils on a diameter of 252 mils or a permanent strain of about 1.2 percent. From Figure 3-4, the maximum calculated cladding temperature was 1830°F.

High temperature tensile tests were run on samples of the same heat of tubing used to clad the fuel specimens. These data are presented in Figures 4-6 and 4-7. From Figure 4-7 it is seen that the stress corresponding to a strain of 0.012 inch/inch is about 8950 psi.

Assuming an effective stress corresponding to a strain of 1.2 percent, the elastic displacement expression for a closed-end cylinder subjected to an internal pressure can be employed:

$$\delta = \frac{R}{E} (\sigma_n - \nu \sigma_z), \quad (24)$$

where

$$\begin{aligned} \delta &= \text{Radial displacement, inch;} \\ R &= \text{Radius, 0.126 inch;} \\ E &= \text{Modulus of elasticity, psi;} \\ \sigma_n &= \text{Circumferential stress, psi;} \\ \sigma_z &= \text{Axial stress, psi; and} \\ \nu &= \text{Poisson's ratio = 0.26.} \end{aligned}$$

$$\text{Since strain} = \frac{\bar{\sigma}}{E},$$

then

$$\frac{\delta}{R} = \frac{\bar{\sigma}}{E}$$

or

$$\delta = \frac{R \bar{\sigma}}{E}.$$

Thus,

$$\frac{R \bar{\sigma}}{E} = \frac{R}{E} (\sigma_n - \nu \sigma_z)$$

or

$$\bar{\sigma} = (\sigma_n - \nu \sigma_z),$$

where

$$\bar{\sigma} = \text{Effective stress.}$$

Substituting

$$\frac{PR}{t} \text{ for } \sigma_n$$

and

$$\frac{PR}{2t} \text{ for } \sigma_z$$

yields

$$\bar{\sigma} = \frac{PR}{t} \left(1 - \frac{\nu}{2} \right),$$

from which

$$P = \frac{\bar{\sigma} t}{R \left(1 - \frac{\nu}{2} \right)},$$

where

t = Wall thickness, 0.015 inch.

Solving for P:

$$P = \frac{(8950)(0.015)}{(0.126) \left(1 - \frac{0.26}{2}\right)},$$

$$P = 1225 \text{ psi.}$$

From previous calculations⁽¹⁰⁾ it is known that the NaK cover gas pressure in the capsule increases to about 440 psig during the transient. Thus, the internal pressure required to cause the 3-mil cladding deformation is 1225 + 440 or 1665 psig.

Although the agreement between the two calculations for internal pressure is something less than precise (1277 psig versus 1665 psig), it appears that fission gas release and cladding deformation can be correlated. From the calculations it is apparent that even though the steady-state gas release model is rather crude, its use results in estimation of gas release during the transient that seems to yield at least a first order correlation of the experimental data.

The basis for the modification of the fission gas model cited by O'Neill,⁽²⁰⁾ may be somewhat nebulous since the gas release measured in the C3A experiment (15 percent) is low in comparison to most of the gas release data obtained in previous irradiations of mixed oxide fuel specimens.⁽²⁵⁾ The calculated internal pressure during the transient is sensitive to not only the gas released during the transient but also to the gas released during the steady-state irradiation since it is this gas which determines the initial pressure and the amount of gas which will be released during the transient for a given radius of molten fuel. Thus, small differences in the amount of

steady-state gas release could alter the calculated transient gas release and internal pressure calculations significantly. This could, of course, very easily account for the differences in the results of the calculations just presented. Further, the precise strength of the cladding and its effective wall thickness were not well known, and these are direct influences on the calculated pressure for deformation. For example, as seen from Figure 3-24(d), cracks extended into the cladding for about 40 percent of its thickness. If the effective wall thickness was reduced accordingly, then the pressure required to deform the cladding would have been 1175 psi (including the 440-psig external pressure term).

4.2 TRANSIENT PERFORMANCE OF IRRADIATED FUEL WITH RESPECT TO A POSTULATED CREDIBLE ACCIDENT IN AN FCR (SPECIMEN C3E)

It will be recalled that the purpose of the C3E experiment was to attempt to duplicate the conditions of radial temperature profile in the fuel and cladding temperature that were typical of postulated credible accidents in an FCR. The need for these experiment conditions was, of course, predicated on the necessity to investigate the transient performance of highly irradiated fuel in light of the results attained in the C3B experiment, viz., the observed cladding deformation and fuel redistribution. Application of the transient gas release model just derived to the design of the transient for the C3E experiment indicated that melting of fuel to a radius of about 60 mils (≈ 30 percent of the fuel volume) would not be expected to result in cladding deformation or significant fuel relocation. This is considerably more melting than anticipated from current analyses of postulated credible accidents in an FCR. (Transient limits given by Cohen and O'Neill⁽⁷⁾ state that no fuel will become completely molten during a postulated credible accident in an FCR.) As discussed in subsection 3.2.3 and illustrated

FIGURE 4-6. STRESS-STRAIN DIAGRAM FOR TYPE 347 STAINLESS STEEL AT 1400°F

FIGURE 4-7. STRESS-STRAIN DIAGRAM FOR TYPE 347 STAINLESS STEEL AT ~1850°F

in Figures 3-30 and 3-35, melting of the fuel to a radius of 51 mils (≈ 22 percent of the volume) took place during the transient irradiation without the occurrence of cladding deformation or significant axial fuel relocation. The most significant point here is that regardless of the accuracy or validity of the fission gas release models employed, it definitely appears that at least some melting in axially unrestrained fuel can be tolerated (on a one-time basis) during an accident.

A comparison of calculated radial temperature profiles from the C3E experiment and from FORE⁽²⁶⁾ - computed fuel temperatures in a \$0.20/sec reactivity addition accident in an FCR - is given in Figure 4-8. The point illustrated is that it is possible to simulate reasonably, in TREAT, the fuel and cladding temperatures calculated to occur in a postulated credible accident in an FCR. The FORE calculation in this case more nearly represents a hypothetical accident since an excursion of this duration would have been terminated by actuation of the control system much before the fuel had attained the indicated temperatures.

4.3 TRANSIENT PERFORMANCE LIMIT OF HIGHLY IRRADIATED FUEL (SPECIMEN C3C)

In reviewing the data from the C3E experiment prior to specifying the test conditions (transient magnitude) for the C3C experiment it was noted that the fission gas release in the C3E experiment was quite high (80 to 93 percent).

Estimation of the fission gas release on the basis of the model cited by O'Neill and modified to fit the C3A data resulted in calculated release values very much lower than those observed in the C3E experiment. On this basis it was decided to revert to the original model for the calculations required to specify the test transient for the C3C experiment. As seen from the analysis of the test data given

in Appendix D, the results of the experiment and the calculations appear to be consistent. One major uncertainty in this experiment was whether or not the steady-state released fission gas escaped during the attempt to measure the gas pressure prior to the transient irradiation. On the basis of the calculations in Appendix D, the observed cladding deformation, and the fact that a large gas activity was not noted in the RML off-gas at the time the attempt to puncture the fuel pin diaphragm was made, it appears reasonable to conclude that the diaphragm was intact at the time of transient irradiation. Since the neutron radiograph (Figure 3-9) indicated the presence of fuel on the diaphragm, it could be postulated that as a combination of pressure and high temperature (which would occur at about the same time as the maximum fuel temperature) the diaphragm failed, allowing molten fuel to be expelled from the pin. The point is that failure of the diaphragm probably occurred at about the same time that the maximum cladding deformation had occurred.

When the results of the transient irradiation of the two high burnup specimens (C3E and C3C) are compared, it becomes obvious that the extent of fuel melting during the transient is a prime factor in predicting transient performance. The extent of fuel melting which occurs is directly related to the transient magnitude. The melting point of the fuel which can change as a result of burnup and plutonium segregation is of lesser importance since the possible shift in melting point is well within the range of experimental error involved in in-pile transient testing of fuel and the transient heat transfer calculations.

Another major factor in determining or predicting transient performance of fuel is the steady-state fission gas release which occurs. This is principally dependent on the steady-state operating conditions; high fuel temperatures will result in higher fission gas release than will low fuel temperatures.

FIGURE 4-8. COMPARISON OF TRANSIENT EXPERIMENT RESULTS WITH CALCULATED FAST CERAMIC REACTOR TEMPERATURE PROFILE DURING $0.20 / \text{sec}$ EXCURSION

A third major factor affecting fuel-cladding transient performance is the strength of the cladding. In these experiments it was observed that cracking of the cladding occurred at the inner surface of which had suffered heavy grain boundary precipitation. The cause of the grain boundary precipitation is not understood. One possibility is that the fuel could have in some way enhanced carburization of the inner surface. Another is sensitization of the cladding through cold working or time-at-temperature. It should be noted however, that the inner surface temperature of the cladding was on the order of 800°F during the steady-state irradiation. Radiation embrittlement is, of course, another possible explanation for the lack of ductility demonstrated by the cladding.

On the basis of the foregoing, it appears that the threshold for transient cladding failure with axially unrestrained fuel can be defined by:

- a. Cladding deformation of 1 percent or less.
- b. Melting of fuel to a radius that extends into a region of the fuel which has not released a significant portion of the fission gas generated in it.

4.4 THERMAL VERSUS FAST FLUX IRRADIATION PERFORMANCE

Since it appears that the transient performance of irradiated oxide fuel is highly dependent on the steady-state gas release, an analysis was made to evaluate possible differences in this regard arising from irradiation in a thermal and a fast flux. The fission gas generation in a fast and a thermal flux is compared in Figure 4-9. From Figure 4-9 it is apparent that in a thermal flux less fission gas will be released during steady-state irradiation for a given fuel temperature than in a fast flux. Thus, during a transient, more

fuel can be melted in the fast flux case before significant transient gas release, and hence high internal pressures occur. To illustrate the point further, the case of a reference FCR fuel rod⁽⁷⁾ irradiated to 100,000 MWd/Te was examined. From Figure 4-10 it is seen that the steady-state gas release was:

$$\begin{array}{rcl}
 \text{From the 100 percent zone:} & & 72 \text{ percent} \\
 \text{From the 50 percent zone:} & & \\
 \frac{81-72}{2} & = & 4.5 \text{ percent} \\
 \text{From the 4 percent zone:} & & \\
 (100-81)(0.04) & \cong & 1 \text{ percent} \\
 \text{Total steady-state release} & & 77.5 \text{ percent} \\
 & \cong & 78 \text{ percent}
 \end{array}$$

In the reference design this gas is vented to the coolant so that gas pressure during a transient originates from the gas still contained in the fuel. Calculations for gas release as a function of percent of molten fuel, gas pressure as a function of percent release, and cladding deformation as a function of pressure and temperature, show that deformation of the cladding will occur once fuel melting progresses beyond the 100 percent release zone. In this case, about 72 percent of the fuel could become molten before the initiation of cladding deformation due to gas pressure. FORE calculations show, however, that at this percentage of molten fuel, the center temperature of the fuel is rapidly approaching the point where significant fuel vapor pressure is being generated, so that it appears that the two failure mechanisms are being approached simultaneously in this case. The analysis did not consider the possibility of cladding deformation due to axial restraint of the fuel thermal expansion and the ΔV on melting since it was only intended to illustrate fission gas release during a transient. This, of course, represents a failure mechanism which might, under the right conditions, occur earlier or about the same time as fission gas pressure and fuel vapor pressure.

FIGURE 4-9. GAS GENERATION IN FAST AND THERMAL FLUXES

FIGURE 4-10. FISSION GAS RELEASE IN REFERENCE FCR FUEL

By comparison, if a vented-to-coolant thermal flux case is examined for the same rod power and fuel surface temperature, the fuel center temperature in the thermal flux case is substantially lower than in the fast flux case. This, in addition to the thermal flux gas generation profile, substantially reduces the steady-state fission gas release. Thus, less molten fuel can be tolerated before the onset of cladding deformation in a transient. It should also be noted that fission gas formation is higher in a thermal flux than in a fast flux,⁽²⁷⁾ which further reduces the amount of fuel melting that can be tolerated during a transient.

4.5 FISSION PRODUCT SWELLING IN HIGH BURNUP FUEL

As mentioned in subsection 3.2.1, the neutron radiographs disclosed that fuel column elongation or growth had occurred during the steady-state irradiation. Further, post-irradiation examination of specimen C3E revealed that the fuel-cladding gap had been closed. If it is assumed that these increases in the fuel dimensions were due to fission product swelling, then an upper limit on fuel swelling can be calculated. An upper limit is specified since, as can be seen from the neutron radiographs (Figures 3-8 and 3-9), the axial elongation includes pellet interface gaps.

4.5.1 Swelling in Specimen C3E

From knowledge of the original dimensions of the fuel, the initial fuel volume was 0.229 in.³. From measurements made on the neutron radiographs and considering sintering and center void formation, the fuel volume after steady-state irradiation was 0.237 in.³. The percentage change in volume was

$$\Delta V = \frac{0.237 - 0.229}{0.229} (100) = 3.49 \text{ percent.}$$

From Table 3-1, burnup was 70,000 MWd/Te, which is equivalent to 7.49 atom percent burnup

or 1.76×10^{21} fissions/cm³ of fuel. Thus, swelling was

$$\frac{3.49 \text{ percent } \Delta V}{7.49 \text{ atom percent}}$$

$$= 0.466 \text{ percent } \Delta V / \text{atom percent burnup,}$$

or

$$\frac{3.49 \text{ percent } \Delta V}{1.76 \times 10^{21} \text{ fissions/cm}^3}$$

$$= 0.198 \text{ percent } \Delta V / 10^{20} \text{ fissions/cm}^3.$$

4.5.2 Swelling in Specimen C3C

$$\text{Original fuel volume} = 0.222 \text{ in.}^3$$

$$\text{Final fuel volume} = 0.229 \text{ in.}^3$$

$$\Delta V = \frac{(0.229 - 0.222)}{0.222} (100) = 3.15 \text{ percent.}$$

$$\begin{aligned} 73,000 \text{ MWd/Te} &= 7.8 \text{ atom percent burnup} \\ &= 1.88 \times 10^{21} \text{ fissions/cm}^3. \end{aligned}$$

$$\frac{3.15 \text{ percent}}{7.8 \text{ atom percent}}$$

$$= 0.404 \text{ percent } \Delta V / \text{atom percent burnup,}$$

or

$$\frac{3.15 \text{ percent}}{18.8 \times 10^{20} \text{ fissions/cm}^3}$$

$$= 0.168 \text{ percent } \Delta V / 10^{20} \text{ fissions/cm}^3.$$

It is surprising to note that these numbers tend to agree with the Bettis data for solid fission product swelling. However, they are substantially lower than the total swelling values of $0.7 \Delta V / 10^{20}$ fissions/cm³ quoted by Bettis. Further, as mentioned previously, the ΔV values include unrecovered thermal expansion so in actuality the true swelling is, as would be expected, probably substantially lower than the values calculated here.

APPENDIX A

FUEL SPECIMEN AND CAPSULE DETAILS

A.1 SERIES III CAPSULE DESIGN

The program for transient irradiation of 0.250-inch-diameter mixed oxide fuel specimens in TREAT involved both nonpre-irradiated (Series II)⁽⁴⁾ and steady-state pre-irradiated (Series III) fuel specimens. To minimize differences in nuclear characteristics, effect economies in design and fabrication, and permit direct comparison of results between nonpre-irradiated and pre-irradiated specimens, all of the capsules were built to the same basic design.

A.1.1 Design Criteria

1. An external capsule configuration that would permit installation in the TREAT reactor core by substitution for a TREAT fuel element.
2. Sufficient heat capacity to absorb all the heat generated by the fuel specimen during transient irradiation in TREAT.
3. Calorimeter-type construction so that heat generated by the fuel specimen during transient irradiation may be measured accurately by temperature rise.
4. Heat transfer characteristics such that steady-state irradiations at desired heat generation rates would not result in excessive fuel and cladding temperatures.
5. An external capsule configuration that would permit steady-state irradiation in a test reactor pool position.
6. Sufficient strength and integrity to sustain internal pressures and provide containment for fuel and other capsule materials.

A.1.2 Capsule Design

The manner in which these criteria were satisfied for the steady-state irradiations in the GETR is depicted by the capsule illustrated in Figure A-1.

Shown in Figure A-2 is the reencapsulation of the steady-state irradiation capsule (inner capsule assembly) into the TREAT capsule whose configuration approximates that of a TREAT fuel element. Reencapsulation was, of course, performed remotely in the RML hot cells.

The capsule design also permitted the remote installation of a pressure transducer to monitor the fission gas pressure in the fuel pin during a transient. As seen in Figure A-3, the fuel pin incorporated a diaphragm at the bottom of the fuel column. By means of a bellows and hypodermic needle arrangement, which is illustrated in Figure A-4, it was possible to puncture the diaphragm and expose the transducer to the fuel pin internal pressure. In Figure A-5 are photographs taken during the remote installation of a pressure transducer on capsule C3B.

A.2 SERIES III FUEL SPECIMEN DESIGN AND FABRICATIONA.2.1 Design

Fuel specimen mechanical design was predicated on the simulation of the then-current FCR reference fuel pin design.⁽⁵⁾

In this regard, fuel pellet density and diameter, cladding diameter and thickness, and fuel-volume-to-fission-gas-plenum-volume ratio duplicate the reference design. In addition, means for installing a pressure transducer on the fuel pin after completion of the steady-state irradiation also had to be provided. Shown in Figure A-3 are the details of the fuel pin construction. The diaphragm at the bottom of the pin permitted the installation of a pressure transducer which was previously equipped with a hypodermic needle to puncture the diaphragm.

The salient as-fabricated data for each fuel specimen are presented in Table A-1.

A.2.2 Fabrication

A.2.2.1 Fuel Fabrication

A 270-gram batch (B-4) of mixed oxide powder was prepared for the Series III specimens. The feed solution contained 28 gm/liter plutonium, 72 gm/liter natural uranium, and ~1 M nitric acid (isotopic analysis of the feed solution is given in Table A-2). Precipitation was accomplished by addition of

ammonium hydroxide to a final pH of 7 to 8. The coprecipitate was stirred for 15 minutes at ambient temperature, and then filtered. The cake was washed three times with 0.1 M NH_4OH , and dried with air and acetone. The reduction cycle consisted of heating to 750°C, holding at 750 to 780°C for 3 hours, and cooling to room temperature in flowing 6 percent hydrogen - 94 percent helium. The reduced power was ball-milled for 7-1/2 hours to provide a -325 mesh powder for pressing.

Pressing and sintering data for the Series III pellets are given in Table A-3.

A.2.2.2 Fuel Pin Component Fabrication

As seen in Figure A-3, all the components for the fuel pins were made from austenitic stainless steel, with the cladding specified as Type-347. Inspection of the cladding consisted of ultrasonic testing for wall defects and burst tests of sample pieces, as well as dimensional measurements. The burst pressure of the three samples tested was ~11,200 psig, at room temperature.

**FIGURE A-2. TREAT CAPSULE ASSEMBLY
FOR TRANSIENT IRRADIATION
OF PRE-IRRADIATED
SPECIMENS**

FIGURE A-3. FUEL SPECIMEN DETAILS

TABLE A-1

DATA FOR SERIES 3 FUEL PINS

Treat Series Number	3-A	3-B	3-C	3-D	3-E
Powder Source - Batch No.	B-4	B-4	B-4	B-4	B-4
(a) Ratio Pu/Pu + U in Mixed Oxide (percent)	28.1	28.1	28.1	28.1	28.1
Inside Diameter of Cladding (inch)	0.221	0.221	0.221	0.221	0.221
Maximum Diameter Range of Pellets (inch)					
Uranium Oxide	0.2157-0.2163	0.2158-0.2161	0.2158-0.2162	0.2157-0.2160	0.2157-0.2161
Mixed Oxide	0.215 -0.2195	0.215 -0.2185	0.215 -0.219	0.2155-0.2195	0.216 -0.2195
Average Diameter of Pellets (inch)					
Uranium Oxide	0.2159	0.2160	0.2161	0.2158	0.2160
Mixed Oxide	0.217	0.2165	0.217	0.2175	0.2185
Diametrical Gap (inch)					
Range	0.002 -0.006	0.003 -0.006	0.002 -0.006	0.002 -0.006	0.002 -0.005
Average	0.004	0.0045	0.004	0.0035	0.0025
Fuel Length (inches)	6.013	6.010	6.0125	6.0375	6.110
Fuel Weight (gm)	38.51	38.84	38.82	38.90	39.525
Fuel Density (Percent of Theoretical)					
Range	93.9-96.9	94.4-97.8	94.5-97.2	94.1-97.2	94.3-97.5
Average	95.0	96.8	96.0	95.2	95.0
(b) Fuel Oxygen/Metal Ratio	1.979 $\pm$ 0.006	1.979 $\pm$ 0.006	1.979 $\pm$ 0.006	1.979 $\pm$ 0.006	1.979 $\pm$ 0.006
Total Weight of Fuel Pin (gm)	74.63	75.20	75.10	75.31	75.88
Date Loaded	8/22/62	8/22/62	8/22/62	8/30/62	8 30 62

(a) Ratio of Pu/Pu + U is based upon plutonium and uranium analyses of sintered pellets. Precision of plutonium and uranium analyses is ± 1 percent; therefore, the precision of the calculated ratio is ± 2 percent (i.e., 20.0 percent is 19.6 to 20.4 percent).

(b) Fuel oxygen/metal ratio was determined for reference pellets from the batch (B-4). Determination was made by technique established by W. L. Lyon and described in a report entitled "The Measurement of Oxygen-to-Metal Ratio in Solid Solutions of Uranium and Plutonium Dioxides," GEAP-4271.

TABLE A-2

ISOTOPIC ANALYSIS OF PLUTONIUM FEED SOLUTION FOR SERIES III FUEL PELLETS

Isotope	Atom Percent
Pu-239	91.60
Pu-240	7.65
Pu-241	0.72
Pu-242	0.03

**FIGURE A-4. PRESSURE TRANSDUCER
INSTALLATION TREAT
SERIES III CAPSULES**

NEG. NO. 31397

(a) REMOVING CAP FROM BOTTOM OF FUEL PIN

NEG. NO. 31400

(c) INSERTION OF HYPODERMIC NEEDLE

NEG. NO. 31398

(b) BOTTOM OF FUEL PIN AFTER CAP REMOVED

NEG. NO. 31401

(d) COMPLETED WELDMENTS

FIGURE A-5. STEPS IN THE REMOTE INSTALLATION OF PRESSURE TRANSDUCER ON SPECIMEN C3B

TABLE A-3

PRESSING AND SINTERING DATA FOR SERIES III PELLETS

(28 percent Plutonium - 72 percent Uranium Mixed Oxide)

Pellet Numbers	Pelletizing Pressure (psig)	Green Density (percent of theoretical)		Sintering		Sintered Density (percent of theoretical)		Firing Shrinkage (percent)		Comments
		Range	Average	Temperature (°C)	Time (h)	Range	Average	Diametric	Axial	
1-4	9,500	46.4-46.6	46.5	1580-1600	4-1/2	95.4-96.5	96.0	22.3	21.4	
5-8	11,000	47.4-47.9	47.7	1580-1600	4-1/2	95.9-97.1	96.7	21.7	20.7	
9-12	14,000	48.4-48.6	48.5	1580-1600	4-1/2	95.9-96.3	96.1			
13-16	20,000	51.1-51.5	51.4	1580-1600	4-1/2	96.0-96.3	96.1	19.6	18.8	
17-20	10,000	46.6-46.8	46.8	1580-1610	4-1/2	94.3-95.1	94.7	21.8	20.8	
21-30	11,000	47.0-47.6	47.3	1580-1610	4-1/2	93.9-95.2	94.5	21.5	20.3	
31-34	12,000	48.5	48.5	1580-1610	4-1/2	94.2-95.4	94.8	21.1	19.7	
35-37	18,000	50.5-50.8	50.6	1580-1610	4-1/2	93.6-94.6	94.2	19.7	18.3	
38-40	23,000	52.8-53.0	52.9	1570-1620	5	92.3-92.9	92.5	18.0	16.7	Warped. (a)
41-42	16,500	49.8-50.1	49.9	1570-1620	5	91.4-92.9	92.2	19.0	18.9	Warped.
44-64	10,500	46.7-48.0	47.3	1570-1620	5	89.9-92.2	91.4	20.5	19.6	Warped and chipped.
65-66	19,500	50.2-50.8	50.5	1560-1620	4	91.5-92.0	91.8	19.2	17.5	Warped and chipped.
67-68	13,500	47.6-50.0	48.8	1560-1620	4	90.0-91.5	90.8	19.6	18.3	Warped.
69-74	11,000	44.7-47.1	46.3	1560-1620	4	88.8-94.6	91.4	21.3	19.8	Warped and chipped.
75-89 ^(b)	11,000	47.5-48.6	47.9	1560-1600	4	96.3-97.8	97.3	22.0	20.7	
90-99 ^(c)	11,000	47.7	47.7	1560-1590	4	95.9-97.0	96.5	22.0	20.5	
100-120 ^(b)	11,000	48.1-48.4	48.2	1560-1590	4	92.3-97.2	94.9	21.2	20.1	
121-144 ^(d)	11,000	47.6-48.3	47.8	1560-1590	4	93.0-95.7	94.5	21.2	19.8	
145-168 ^(e)	11,000	47.7-48.3	48.0	1570-1600	4	93.2-95.3	94.7	21.1	20.2	
169-177	10,500	46.8-48.0	47.5	1480-1530	4	96.2-97.5	96.8	22.0	20.9	

NOTES:

- (a) Hour-glass configuration.
 (b) Green data for three pellets.
 (c) Green data for two pellets.
 (d) Green data for seven pellets.
 (e) Green data for five pellets.

APPENDIX B

COMPARISON OF MEASURED FISSION GAS RELEASE TO
CALCULATED FISSION GAS RELEASE FOR SPECIMEN C3A

From the interim report,⁽¹⁰⁾ the measured fission gas release in specimen C3A was 15.5 percent. Since the fuel composition and capsule construction was identical to that for specimen C3B, the power profile and fission gas generation curves given in Figures 4-1 and 4-2 are also applicable to specimen C3A.

For determination of the radial temperature profile in the fuel, $\int k dt$ curve given in Figure 4-3 and $\int_{T_s}^{T_r} k dT$ calculations for a linear power generation of 20.4 kW/ft were used to obtain the curve given in Figure B-1.

Determination of the fission gas release according to the model cited by O'Neill and from Figure B-1 is made as follows:

At $T = 3272^\circ\text{F}$ (1800°C), $r = 0.083$ inch.
For $r = 0.083$ inch, the fission gas content is 45.5 percent.

At $T = 2912^\circ\text{F}$ (1600°C) $r = 0.0895$ inch.
For $r = 0.0895$ inch, the fission gas content is 49.5 percent.

Gas release from 100 percent release zone ($T \geq 1800^\circ\text{C}$) = 45.5 percent.

Gas release from 50 percent release zone ($1600^\circ\text{C} \leq T \leq 1800^\circ\text{C}$) = (0.5) (49.5-45.5) = 2 percent.

Gas release from 4 percent release zone ($T \leq 1600^\circ\text{C}$) = (0.04) (100-49.5) = 2 percent.

Total steady-state gas release = 45.5
+ 2 + 2 = 49.5 percent.

This is substantially higher than the measured 15 percent. Since the release zones are rather coarse, 100, 50, and 4 percent, it may be better to assume that 100 percent release occurs at a higher fuel temperature. By forcing the model to yield a 15.6 percent release value, the temperature at which the various release are assumed to occur can be determined by a trial and error method.

Assume 100 percent for $T \geq 4200^\circ\text{F}$
50 percent for $3900^\circ\text{F} \leq T \leq 4200^\circ\text{F}$
4 percent for $T \leq 3900^\circ\text{F}$

From Figure B-1:

At $T = 4200^\circ\text{F}$, $r = 0.056$ inch, and the gas content is 12.5 percent.

At $T = 3900^\circ\text{F}$, $r = 0.068$ inch, and the gas content is 21.8 percent.

Gas release from 100 percent zone = (1.0) (12.5) = 12.5 percent

Gas release from 50 percent zone = (0.5) (21.8-12.5) = 4.7 percent

Gas release from 4 percent zone = (0.04) (100-21.8) = 3.1 percent
Total 20.3 percent

Assume 100 percent for $T \geq 4300^\circ\text{F}$
50 percent for $4000^\circ\text{F} \leq T \leq 4300^\circ\text{F}$
4 percent for $T \leq 4000^\circ\text{F}$

At $T = 4300^\circ\text{F}$, $r = 0.05$ inch, and the gas content is 9 percent.

At $T = 4000^\circ\text{F}$, $r = 0.0645$ inch, and the gas content is 18.5 percent.

FIGURE B-1. SPECIMEN C3A RADIAL TEMPERATURE PROFILE AND FISSION GAS GENERATION

Gas release from 100 percent
 zone = (1.0) (9) = 9 percent

Gas release from 50 percent
 zone = (0.5) (18.5-9) = 4.75 percent

Gas release from 4 percent
 zone = (0.04) (100-18.5) = 3.25 percent
 Total 17.01 percent

Assume 100 percent release for $T \geq 4350^{\circ}\text{F}$
 50 percent release for $4000^{\circ}\text{F} \leq T \leq 4350^{\circ}\text{F}$
 4 percent release for $T \leq 4000^{\circ}\text{F}$

At $T = 4350^{\circ}\text{F}$, $r = 0.046$ inch, and the gas content is 7.2 percent.

At $T = 4000^{\circ}\text{F}$, $r = 0.0645$ inch, and the gas content is 18.5 percent

Gas release from 100 percent
 zone = (1.0)(7.2)

Gas release from 50 percent
 zone = (0.5) (18.5-7.2) = 5.7 percent

Gas release from 4 percent
 zone = (0.04) (100-18.5) = 3.3 percent
 Total 16.2 percent

Assume 100 percent for $T \geq 4400^{\circ}\text{F}$
 50 percent for $4100^{\circ}\text{F} \leq T \leq 4400^{\circ}\text{F}$
 4 percent for $T \leq 4100^{\circ}\text{F}$

At $T = 4400^{\circ}\text{F}$, $r = 0.032$ inch, and the gas content is 2 percent.

At $T = 4100^{\circ}\text{F}$, $r = 0.06$ inch, and the gas content is 15 percent.

Gas release from 100 percent
 zone = (1.0) (2) = 2 percent

Gas release from 50 percent
 zone = (0.5) (15-2) = 6.5 percent

Gas release from 4 percent
 zone = (0.04) (100-15) = 3.6 percent
 Total 12.1 percent

From the foregoing it is apparent that the correct release value can be obtained with 100 percent release occurring at a temperature between 4300 and 4350°F . For this analysis, it shall be assumed that 100 percent release occurs at $T \geq 4350^{\circ}\text{F}$, 50 percent release occurs at $4000^{\circ}\text{F} \leq T \leq 4350^{\circ}\text{F}$, and 4 percent release occurs at $T \leq 4000^{\circ}\text{F}$.

APPENDIX C

ESTIMATE OF FISSION GAS RELEASE FROM SPECIMEN C3E

C.1 FISSION GAS RELEASE DURING STEADY-STATE IRRADIATION

From the interim report, ⁽¹⁰⁾ the average rod power during the last cycle of the steady-state irradiation was 20.2 kW/ft. In this case, this was also the highest average rod power achieved with the exception of the first cycle of irradiation. Thus, the gas release estimate will reflect the increased fuel temperatures during the last cycle of irradiation.

The radial temperature distribution in the fuel was determined by calculating the $\int_{T_s}^{T_r} k dT$ to determine the local radial temperatures. This was accomplished in the following manner.

C.1.1 Fuel Surface Temperature

From the interim report, Appendix B, page B-6, the sum of all the temperature drops from the fuel surface to the capsule wall was 2210°F for a rod power of 29.3 kW/ft. By ratio, for 20.2 kW/ft, this becomes $\left(\frac{20.2}{29.3}\right) (2,210) = 1,523^\circ\text{F}$.

For a coolant temperature of 110°F, the fuel surface temperature is $T_s = \Sigma\Delta T + T_w$
 $= 1523 + 110 = 1633^\circ\text{F}$ (890°C) ($h_{\text{fuel-cladding}}$
 $= 1500 \text{ Btu/h-ft}^2\text{-}^\circ\text{F}$).

C.1.2 Radial Temperature Profile

From physics calculations, the power generation profile was determined to be as shown in Figure C-1. This curve was fitted by a sixth-power polynomial equation with a

least-squares computer routine. The coefficients are then used in another computer

program which solves the $\int_{T_s}^{T_r} k dT$ equation given on page 135. Once the $\int_{T_s}^{T_r} k dT$ has

been determined from the computer calculations, the temperature at any radius is determined from the $\int k dT$ curve for mixed oxide fuel ⁽²¹⁾ given in Figure 4-3 and the expression

$$\int_0^{T_r} k dT = \int_0^{T_s} k dT + \int_{T_s}^{T_r} k dT.$$

Figure C-2 presents the calculated radial temperature profile in the fuel.

C.1.3 Fission Gas Generation and Distribution

If it is assumed that fission gases are generated in direct proportion to the power generation in the fuel, then fission gas generation as a function of the fuel radius is obtained by integration of the power distribution curve:

$$\text{Percent gas} = \frac{\int_a^r f(r) dr}{\int_a^b f(r) dr} \quad (100),$$

where

$$f(r) = 0.4054 + 0.2767r + 38.19r^2 - 1333r^2 + 34,460r^4 - 293,500r^5 + 879,400r^6.$$

a = inner fuel radius

b = outer fuel radius

The results of this calculation are presented in Figure C-3.

FIGURE C-1. CALCULATED POWER PROFILE IN SPECIMEN C3E

FIGURE C-2. CALCULATED STEADY-STATE TEMPERATURE PROFILE IN SPECIMEN C3E AT END OF IRRADIATION

FIGURE C-3. FISSION GAS GENERATION IN SPECIMEN C3E

$$\int_{T_b}^{T_r} k dT = \left(\frac{Q'_b}{2\pi} \right) \frac{\left\{ \frac{H}{4} (b^2 - r^2) + \frac{J}{9} (b^3 - r^3) + \frac{K}{16} (b^4 - r^4) + \frac{L}{25} (b^5 - r^5) + \frac{M}{49} (b^6 - r^6) + \frac{N}{49} (b^7 - r^7) + \frac{P}{64} (b^8 - r^8) + \ln \frac{r}{b} \left[\frac{Ha^2}{2} + \frac{Ja^3}{3} + \frac{Ka^4}{4} + \frac{La^5}{5} + \frac{Ma^6}{6} + \frac{Na^7}{7} + \frac{Pa^8}{8} \right] \right\}}{\left[\frac{H}{2} (b^2 - a^2) + \frac{J}{3} (b^3 - a^3) + \frac{K}{4} (b^4 - a^4) + \frac{L}{5} (b^5 - a^5) + \frac{M}{6} (b^6 - a^6) + \frac{N}{7} (b^7 - a^7) + \frac{P}{8} (b^8 - a^8) \right]}$$

where

Q'_b = Rod power, Btu/h-ft;

$$H = \left(1 - \frac{B}{\alpha} a + \frac{C}{\alpha} a^2 - \frac{D}{\alpha} a^3 + \frac{E}{\alpha} a^4 - \frac{F}{\alpha} a^5 + \frac{G}{\alpha} a^6 \right);$$

$$J = \left(\frac{B}{\alpha} - \frac{2C}{\alpha} a + \frac{3D}{\alpha} a^2 - \frac{4E}{\alpha} a^3 + \frac{5F}{\alpha} a^4 - \frac{6G}{\alpha} a^5 \right);$$

$$K = \left(\frac{C}{\alpha} - \frac{3D}{\alpha} a + \frac{6E}{\alpha} a^2 - \frac{10F}{\alpha} a^3 + \frac{15G}{\alpha} a^4 \right);$$

$$L = \left(\frac{D}{\alpha} - \frac{4E}{\alpha} a + \frac{10F}{\alpha} a^2 - \frac{20G}{\alpha} a^3 \right);$$

$$M = \left(\frac{E}{\alpha} - \frac{5F}{\alpha} a + \frac{15G}{\alpha} a^2 \right);$$

$$N = \left(\frac{F}{\alpha} - \frac{6G}{\alpha} a \right);$$

$$P = \left(\frac{G}{\alpha} \right);$$

$$\alpha = \text{Ratio of power at center to power at surface} \left(\frac{P_c}{P_s} \right);$$

B, C, D, E, F, and G are coefficients from sixth power polynomial equation for power profile in fuel; and

a, b, r are fuel dimensions.

Annular Fuel Pellet

By superimposing the gas generation and temperature profiles on the same plot and using fuel pellet volume ratio $(r/R)^2$, rather than the radius as an abscissa, the plot shown in Figure C-4 is obtained.

C.1.4 Fission Gas Release

It is known that fission gas release in ceramic fuel is a function of, among other things, temperature. A number of workers^(18, 19, 20) have proposed various percentages of gas release for various temperature zones in the fuel. The model used in this analysis is that cited by O'Neill.⁽²⁰⁾ Thus, gas release is assumed to be related to fuel temperature by:*

100 percent release for $T \geq 1,800^\circ\text{C}$,

50 percent release for $1,600^\circ\text{C} \leq T \leq 1,800^\circ\text{C}$, and

4 percent release for $T \leq 1,600^\circ\text{C}$.

From Figure C-4 it is seen that at a fuel temperature, $T = 3,272^\circ\text{F}$ ($1,800^\circ\text{C}$), the fission gas content is 42.0 percent, and at $T = 2,912^\circ\text{F}$ ($1,600^\circ\text{C}$), the fission gas content is 51.8 percent. The steady-state gas release is then determined by summing the gas release from the various zones:

100 percent release zone
Gas released = $(1.0) (42.0) = 42.0$ percent

50 percent release zone

Gas released = $(0.5) (51.8 - 42) = 4.9$ percent

4 percent release zone

Gas released = $(0.04) (100 - 51.8) = 1.9$ percent

Total Steady-State Gas Release = 48.8 percent

As indicated in Figure C-5, the axial flux profile, and consequently, the power generation in the pin, varies during the cycle. It also is seen that, in general, the lower half of the fuel pin was subjected to a higher flux than the upper portion. Thus, a correction for additional fission gas release is obtained by considering the higher fuel temperatures in the lower portion of the fuel during the start of the last cycle. As seen from Table 3-1, the start-of-cycle peak power for specimen C3E was estimated to be 23.9 kW/ft. By applying the calculational technique just presented for 1-inch lengths of the fuel, it was determined that the additional gas release amounted to 8.1 percent, so that the calculated total gas release becomes 56.9 percent.

C.2 FISSION GAS RELEASE DURING TRANSIENT IRRADIATION

One of the basic assumptions regarding fission gas release during a transient is that gas is released only from fuel which is melted. This assumption is based on the premise that fission gas release from oxide fuel is principally a diffusion process and that in a transient excursion of short duration, there is not sufficient time for extensive gas release by this process.

From the photomicrographs presented in Figure 3-28, it was determined that the molten radius was 0.051 inch, or 22 percent of the fuel was molten. From Figure C-4, it is seen that transient melting of the fuel occurred in a region that had already released

*This is in contrast to the modified model presented in Section IV in the derivation of the transient gas release model from the C3B experimental results. However, the very low gas release measured in specimen C3A which is the basis for the modification of the steady-state gas release model does not appear to be wholly consistent with gas release data from previous mixed oxide fuel specimens.⁽²⁵⁾

FIGURE C-4. STEADY-STATE TEMPERATURE PROFILE AND FISSION GAS GENERATION, SPECIMEN C3E

FIGURE C-5. GETR POOL POSITION AXIAL FLUX SHAPES

all of the gas generated within it during the steady-state irradiation. Thus, in this instance, no gas release is calculated to have occurred during transient irradiation of the fuel.

C.3 RELEASED FISSION GASES VERSUS GENERATED GASES

Estimation of the total atoms of fission gases formed is dependent on the total burnup (total fissions) and the conversion of Xe-135 to Xe-136.

C.3.1 Total Number of Fissions

From Table 3-1, the burnup as estimated from the thermocouple data was 70,000 MWd/Te. The weight of fuel was 39.525 grams of mixed oxide (28.1 percent PuO₂), which was equivalent to 34.82 grams of Pu + U. Assuming 200 MeV/fission and 2.696×10^{21} fissions/MWd results in a total of 6.58×10^{21} fissions.

C.3.2 Conversion of Xe-135 to Xe-136

From the mass spectrometric analysis of the fission gas, Table 3-6, it is seen that Xe-136 accounted for 39.3 percent of the total xenon.

For the fissioning of Pu-239 in a thermal flux, the fission yield for Xe-136 as given by Weaver⁽²⁸⁾ is 6.58 percent, the yield for Xe-135 is 7.02 percent, and the total xenon yield of 30.01 percent. The yield for Xe-136 with respect to the total yield of xenon is $\frac{6.58}{30.01} = 0.219$ or 21.9 percent, which is substantially less than the measured value. The additional Xe-136 is, of course, the result of conversion of Xe-135 to Xe-136. The extent

of the conversion was estimated in the following manner.

$$\frac{\text{Xe-136}}{\text{Total xenon}} = \frac{6.58}{30.01} = 0.219 \text{ or } 21.9 \text{ percent}$$

$$\frac{\text{Xe-135}}{\text{Total xenon}} = \frac{7.02}{30.01} = 0.234 \text{ or } 23.4 \text{ percent}$$

$$\text{Total } 45.3 \text{ percent}$$

If all the Xe-135 were transmuted to Xe-136, the Xe-136 content would have been 45.3 percent. The percent conversion of Xe-135 to Xe-136 is equal to the difference between the maximum possible Xe-136 production (45.3 percent) and the Xe-136 produced (39.3 percent) subtracted from the yield of Xe-135 with respect to the total xenon (23.4 percent) divided by the yield of Xe-135 with respect to the total xenon.

$$\frac{23.4 - (45.3 - 39.3)}{23.4} (100) = 74.4 \text{ percent.}$$

Comparison of this value to the values for Xe-135 conversion obtained in other experiments indicates that the conversion in this case is low. The conversion values calculated from other FCR experiments are presented in Table C-1. It is readily seen that a value of 90 to 95 percent would be more consistent with these data, especially in light of the fact that the range of precision in the C3E measurements (Table 3-6) could very easily account for the difference between the 95 percent and 74.4 percent values. Accordingly, the estimate of fission gas generation will be based on a Xe-135 conversion of 95 percent.

Once again from Weaver,⁽²⁸⁾ the fission yields for krypton and xenon are as given in Table C-2.

TABLE C-1
CONVERSION OF Xe-135 TO Xe-136 IN FCR
FUEL SPECIMEN THERMAL FLUX IRRADIATIONS

<u>Experiment</u>	<u>Xe-136 (percent)</u>	<u>Conversion Xe-135 to Xe-136 (percent)</u>	<u>Reference</u>
C3D	44	94.5	10
E2B	43	90.2	13

TABLE C-2
XENON AND KRYPTON YIELDS FOR
THERMAL FISSION OF U-235 AND Pu-239

<u>Krypton</u>	<u>U-235 (percent Yield)</u>	<u>Pu-239 (percent Yield)</u>	<u>Xenon</u>	<u>U-235 (percent Yield)</u>	<u>Pu-239 (percent Yield)</u>
83	0.5428	0.2901	131	2.9316	2.7581
84	1.0050	0.4670	132	4.3801	5.2071
85	0.3049	0.1215	134	8.0332	7.4422
86	<u>2.0103</u>	<u>0.7582</u>	135*	6.4696	7.0236
			136	<u>6.4123</u>	<u>6.5794</u>
Total Krypton	3.8630	1.6368			
			Total Xenon*	28.2268	30.0104

*Assumes 100 percent conversion to Xe-136.

Since only 95 percent of Xe-135 is assumed to have been converted to Xe-136, the xenon yield for thermal fissioning of U-235 and Pu-239 becomes 27.0933 and 29.6592 percent, respectively.

Krypton Yield

$$(0.014)(3.8630) + (0.986)(1.638) \\ = 1.66915 \text{ percent.}$$

Xenon Yield

$$(0.014)(27.9033) + (0.986)(29.6592) \\ = \underline{29.6346 \text{ percent}} \\ \text{Total Gas Yield} = 31.30 \text{ percent}$$

From Table 3-7, the ratios of U-235 and Pu-239 plus Pu-241 fission to the total number of fissions are:

$$\text{U-235 fissions} = \frac{0.10}{7.18}(100) = 1.4 \text{ percent,}$$

Thus, 31.3 percent of the fissions result in the formation of fission product gases.

and by difference, 98.6 percent of the fissions occurred in the Pu-239 and Pu-241.*

The total number of atoms of fission gases formed is:

*Since the percent of fissions occurring in Pu-241 was extremely small and the fission yields for Pu-241 are quite close to those for Pu-239, the Pu-241 fissions have been combined with the Pu-239 fissions.

$$\left(6.58 \times 10^{21} \text{ fissions}\right) \left(0.313 \frac{\text{atoms}}{\text{fission}}\right) \\ = 2.06 \times 10^{21} \text{ atoms,}$$

which is equivalent to

$$\frac{(2.06 \times 10^{21} \text{ atoms}) \left(2.24 \times 10^4 \frac{\text{cm}^3}{\text{mol}} \right)}{6.023 \times 10^{21} \frac{\text{atoms}}{\text{mol}}}$$

$$= 76.6 \text{ cm}^3 \text{ of gas at STP.}$$

The measured volume of gas in the fuel pin was 69.2 cm^3 at STP (Table 3-5). Correcting for partial pressures of gases other than xenon and krypton:

$$\begin{aligned} \text{Volume of krypton} &= (69.2) (0.05) \\ &= 3.46 \text{ cm}^3 \text{ at STP} \end{aligned}$$

$$\begin{aligned} \text{Volume of xenon} &= (69.2) (0.844) \\ &= 58.40 \text{ cm}^3 \text{ at STP} \end{aligned}$$

$$\text{Total xenon and krypton} = 61.86 \text{ cm}^3 \text{ at STP}$$

The fission gas released based on calculated volume formed and measured volume released then is:

$$\frac{61.86}{76.6} (100) = 80.8 \text{ percent,}$$

which does not agree well with the calculated value of 56.9 percent.

Another means of determining fission gas release is from the ratio of Kr-85 atoms released to Kr-85 atoms formed. From Table 3-5, there were 1.1×10^{17} atoms of Kr-85 per cm^3 of gas. The total gas in the pin was 69.2 cm^3 , so the total number of Kr-85 atoms released was:

$$\begin{aligned} &(1.1 \times 10^{17} \text{ atoms Kr-85/cm}^3)(69.2 \text{ cm}^3) \\ &= 7.612 \times 10^{18} \text{ atoms Kr-85.} \end{aligned}$$

The formation of Kr-85 atoms (atoms Kr-85/fission) was:

$$(0.014) (0.3049) + (0.986) (0.1215)$$

$$= 0.1241 \text{ percent } \frac{\text{atoms}}{\text{fission}}.$$

Atoms of Kr-85 formed were:

$$\begin{aligned} &\left(6.58 \times 10^{21} \text{ fissions} \right) \left(0.001241 \frac{\text{atoms}}{\text{fission}} \right) \\ &= 8.166 \times 10^{18} \text{ atoms.} \end{aligned}$$

Percent fission gas release

$$= \frac{7.612 \times 10^{18} \text{ atoms released}}{8.166 \times 10^{18} \text{ atoms formed}} (100)$$

$$= 93.2 \text{ percent,}$$

which does not disagree too badly with the calculation based on volumes. One explanation for the high measured release as compared to the calculated steady-state value is that as the fuel cools and cracks after a transient, gas is undoubtedly released since fission gas bursts on power changes have been noted in fission gas release measurement experiments. (22, 23)

It is interesting to note that if the steady-state gas release model is applied to the temperature profile given in Figure 3-5, a total fission gas release of 84 percent is predicted. This agrees quite well with the measured release data.

C.4 MICRO-CORE SAMPLE ANALYSES FROM SPECIMEN C3E

To verify the plutonium segregation suggested by the alpha autoradiographs of specimen C3E (Figures 3-30, 3-31, 3-33, and 3-34), micro-core samples were taken from the transverse sections for chemical analyses. The location of the samples is illustrated in Figure C-6, and the dimensional data are presented in Table C-3.

The following report of the work was extracted from a letter report by J. H. Davies who directed the analyses and prepared the report.

C.4.1 Results

The micro-cores were dissolved in hot concentrated nitric acid with small amounts of cerium and cesium carriers added.

Dissolution was very slow for some of the samples. The concentrations of Cs-137 and Ce-144 in the solutions were determined by standard radiochemical methods. The concentrations of uranium in the solutions (in atoms/ml) were determined by mass spectrometry, as were the plutonium and uranium isotopic abundances and the plutonium/uranium ratios. Alpha spectrometry was performed on the solutions from C3E-6E to ascertain the relative contributions of different species to the alpha autoradiographs.

The results of the radiochemical analyses are presented in Tables C-4 through C-8.

C.4.2 Discussion

It may be seen from Table C-3 that the sampling resolution was not very fine, however, the technique virtually eliminates the possibility of cross contamination.

The plutonium isotopic abundances in Table C-4 and the alpha pulse height analyses in Table C-5 both indicate the thermal flux depression through the fuel. The greater burnout of Pu-239 and the increased abundances of higher plutonium and transplutonium isotopes approaching the fuel periphery are evident in both of these sets of data.

From Table C-6 it would appear that some segregation of the plutonium has occurred since there is a definite enrichment of plutonium in the central region of the fuel.

From the data in Tables C-4, C-5, and C-6, the relative specific alpha activities of the C3E-6E micro-cores have been calculated. These are a measure of the relative alpha intensities of the respective regions in the autoradiographs.

The fission product redistribution data are presented in Table C-8. These data were collected as a matter of routine, but interpretation is difficult because of the segregation of the fuel components. In both specimens there is a region immediately adjacent to the bright annulus in the autoradiograph which is depleted in Cs-137. The Cs-137 concentration in the once-molten fuel adjacent to the central void is surprisingly high. Considering the neutron flux depression from the surface to the center of the fuel, Ce-144 appears to be enriched at the center.

Similar effects have been observed in P.A. 10 fuel specimens E3A⁽²⁹⁾ and E1B.⁽³⁰⁾ The segregation mechanism has not been established, but in all cases extensive center melting had occurred.

Interpretation of the C3E results is complicated because of the transient irradiation in TREAT subsequent to the steady-state irradiation in the GETR. The fuel microstructure

NEG. NO. 37105

(a) SAMPLE C3E-6E

NEG. NO. 38010

(b) SAMPLE C3E-8H

FIGURE C-6. MICROCORE SAMPLES FROM SPECIMEN C3E

TABLE C-3

ALPHA RADIOGRAPH STRUCTURE AND MICRO-CORE LOCATION

Fuel Sample	Distance of Radiograph Feature from Fuel Outside Diameter (mils)		Micro-Core Sample	Distance from Fuel Outside Diameter (mils)
	Central Void	Bright Annulus		
C3E-6E	—	64-67	1	73-98
			2	53-68
			3	18-43
			4	18-35
			5*	0-43
C3E-8H	83-86	65-68	1	69-87
			2	41-64
			3	11.5-39
			4	0-18

*This sample was a chip of fuel rather than a micro-core. From the analysis it appears that the location was actually between samples 2 and 3.

TABLE C-4

PLUTONIUM ISOTOPIC ABUNDANCES

Sample		Mass Number			
		Pu-239	Pu-240	Pu-241	Pu-242
C3E-6E	1	0.8021	0.1778	0.0182	0.0018
	2	0.7831	0.1935	0.0210	0.0024
	3	0.7346	0.2337	0.0280	0.0037
	4	0.7340	0.2340	0.0282	0.0038
	5	0.7519	0.2193	0.0256	0.0033
C3E-8H	1	0.8101	0.1709	0.0172	0.0017
	2	0.7926	0.1859	0.0193	0.0021
	3	0.7328	0.2352	0.0280	0.0039
	4	0.6808	0.2744	0.0385	0.0063

TABLE C-5

ALPHA PULSE HEIGHT ANALYSIS

Sample	Ratio of Alpha Peak to That at 5.15 MeV			
	5.15 MeV ^(a)	5.45 MeV ^(b)	5.73 MeV ^(c)	6.01 MeV ^(d)
1	1.0	0.153	0.0041	0.179
2	1.0	0.162	0.0045	0.194
3	1.0	0.194	0.0065	0.261
4	1.0	0.194	0.0068	0.259
5	1.0	0.188	0.0061	0.238

NOTES:

- (a) Pu-239 plus Pu-240
- (b) Pu-238 plus Am-241
- (c) Cm-244
- (d) Cm-242

TABLE C-6

PLUTONIUM-TO-URANIUM RATIO

Sample	Pu-239/U-238	Total Pu*/Total U	Pu/Pu + U
C3E-6E	1	0.395	0.489
	2	0.210	0.268
	3	0.198	0.268
	4	0.190	0.258
	5	0.200	0.264
C3E-8H	1	0.352	0.435
	2	0.222	0.280
	3	0.202	0.276
	4	0.172	0.252
Pre-irradiated	0.356	0.390	0.280
Average at 65,000 MWd/Te (Calculated)		0.287	0.223

*Plutonium isotopic abundances from Table C-4.

Isotopic abundance of U-238 $\approx$ 0.994.

TABLE C-7

RELATIVE ALPHA SPECIFIC ACTIVITIES*

Sample	Relative Specific Alpha Activity
C3E-6E	1
	2
	3
	4
	5

*Calculated from Pu/Pu + U ratios (Table C-6), plutonium isotopic analyses (Table C-4), and alpha pulse height analyses (Table C-5).

TABLE C-8

RADIAL DISTRIBUTION OF Ce-144 AND Cs-137

Sample	Ce-144/U-238	Ce-144/U + Pu	Cs-137/U-238	Cs-137/U + Pu
C3E-6E	1	0.430	0.289	27.4
	2	0.366	0.289	2.5
	3	0.440	0.347	13.2
	4	0.481	0.377	8.8
	5	0.404	0.320	4.5
C3E-8H	1	0.399	0.278	15.9
	2	0.352	0.275	1.8
	3	0.541	0.424	12.7
	4	0.569	0.455	29.6

and evidence of center melting are a consequence of the TREAT irradiation. However, there may be some evidence of segregation occurring during steady-state irradiation as illustrated by the dark annulus surrounding the center void of the fuel in the neutron radiographs, Figures 3-8 and 3-9. This could account for the apparent segregation of the transplutonium elements and cerium which were probably formed in situ in fuel already enriched in plutonium. Further, the segregation which was observed in specimens E3A

and E1B occurred in steady-state GETR irradiations.

The effect of plutonium segregation as observed here on the prompt negative Doppler coefficient has been estimated by Lauritzen, et al., (GEAP-4466), and shown to be small. Another possible effect is the lowering of the melting point of the fuel in the center region which would increase the probability of center melting of high burnup fuel.

ESTIMATION OF INTERNAL PRESSURES IN SPECIMEN C3C

D.1 EXPERIMENTAL DATA

1. Measured cladding deformation was 2.3 mils or 0.91 percent (Table 3-8).
2. Observed molten boundary radius was 90 mils (Figure 3-47).
3. Maximum average cladding temperature was 1295°F.

D.2 CALCULATED FISSION GAS RELEASE

Following the method outlined in the derivation of the transient fission gas release model in Section IV and in Appendix C:

D.2.1 Steady State Fission Gas Release

From Table 3-1, the average rod power during the steady state irradiation was 19 kW/ft. The power profile in the fuel was as given in Figure D-1; the radial temperature profile and the fission gas generation profiles were as given in Figures D-2 and D-3, respectively. From Figure D-2, at $T = 3272^\circ\text{F}$, $r = 0.0795$ inch; at $T = 2912^\circ\text{F}$, $r = 0.086$ inch. From Figure D-3, at $r = 0.0795$ inch, percent gas = 41; at $r = 0.086$ inch, percent gas = 50. The steady-state gas release then was:

$$\begin{aligned}
 &100 \text{ percent Release Zone:} \\
 &\quad (1.0)(41) = 41 \text{ percent} \\
 &50 \text{ percent Release Zone:} \\
 &\quad (0.5)(50-41) = 4.5 \text{ percent} \\
 &4 \text{ percent Release Zone:} \\
 &\quad (0.04)(100-50) = 2.0 \text{ percent} \\
 &\text{Total steady-state} \\
 &\text{gas release} \quad \quad \quad 47.5 \text{ percent}
 \end{aligned}$$

D.2.2 Transient Gas Release

From the microstructure of the fuel, Figure 3-47, the molten boundary radius was 0.090 inch. From Figure D-3, the gas content at 0.090 inch was 57 percent. The transient gas release was:

$$\begin{aligned}
 &50 \text{ percent Release Zone:} \\
 &\quad \quad \quad = 4.5 \text{ percent} \\
 &4 \text{ percent Release Zone:} \\
 &\quad \quad \quad (0.96)(57-60) = 6.7 \text{ percent} \\
 &\text{Total transient} \\
 &\text{gas release} \quad \quad \quad 11.2 \text{ percent}
 \end{aligned}$$

D.2.3 Calculated Internal Pressure due to Fission Gas Release

1. Total Fission Gas Formed during Pre-Irradiation

$$\begin{aligned}
 &\text{Burnup} = 73,000 \text{ MWd/Te} \\
 &(\text{See Table 3-1}).
 \end{aligned}$$

Assume fission gas generation is directly proportional to burnup.

$$\begin{aligned}
 V_{fg} &= (8.71 \text{ cm}^3) \left[\frac{73,000 \text{ MWd/Te}}{8,100 \text{ MWd/Te}} \right] \\
 &\quad \left[\frac{38.82 \text{ gm}}{38.84 \text{ gm}} \right],
 \end{aligned}$$

$$V_{fg} = 78.5 \text{ cm}^3 \text{ at STP}$$

where

$$\begin{aligned}
 8.71 \text{ cm}^3 &= \text{Volume of fission gas generated in specimen C3B (see subsection 4.1.3).} \\
 8100 \text{ MWd/Te} &= \text{Burnup for specimen C3B (see Table 3-1).}
 \end{aligned}$$

38.82 gm = Mass of fuel in specimen C3C (see Table A-1).

38.84 gm = Mass of fuel in specimen C3B (see Table A-1).

Steady-state fission gas release = (47.5 percent) (78.5 cm³),
= 37.3 cm³ at STP.

2. Total Internal Pressure Immediately Prior to Transient

Total internal void volume = 0.232 in. cm³ (see subsection 4.1.3),
= 3.76 cm³.

∴ Helium fill gas volume = 3.76 cm³ at 1 atm and 60°F,
= 3.56 cm³ at STP.

Total gas in pin and pressure transducer assembly after steady-state irradiation = 37.3 + 3.56
= 40.9 cm³ at STP

Pressure at 32°F in fuel pin = (1 atm) $\left[\frac{40.9 \text{ cm}^3}{3.76 \text{ cm}^3} \right]$
= 10.88 atm.

Pressure at preheat of 480°F = (10.88) $\left[\frac{940^\circ\text{R}}{492^\circ\text{R}} \right]$
= 19.1 atm.

3. Maximum Gas Pressure Induced by Transient

Transient fission gas release = (11.2 percent) (78.5 cm³)
= 8.78 cm³ at STP.

Assume that all of the transiently-released fission gas remains within the void volume contained

in the fueled portion of the pin (cladding deformation occurred only in the fueled section of the pin, see subsection 3.2.4 of Results).

Transiently-released fission gas pressure at the standard temperature of 32°F = (1 atm) $\left[\frac{8.78 \text{ cm}^3}{0.28 \text{ cm}^3} \right]$
= 31.3 atm,

but this transiently-released gas is at a temperature of 4900°F (fuel melt temperature).

∴ $P_{\text{tfg}4900} = 31.3 \text{ atm} \left[\frac{5360^\circ\text{R}}{492^\circ\text{R}} \right]$
= 341 atm

Meanwhile the steady-state fission gas and cover gas, trapped in the central void, contribute

$P_{\text{ss}4900} = (10.88 \text{ atm}) \left[\frac{5360^\circ\text{R}}{492^\circ\text{R}} \right]$
= 119 atm

∴ $P_{\text{max}} = P_{\text{tfg}} + P_{\text{ss}} = 341 \text{ atm} + 119 \text{ atm};$
 $P_{\text{max}} = 460 \text{ atm},$
= 6760 psia,
= 6745 psig.

D. 2.4 Calculated Pressure to Cause Observed Deformation

From the derivation given in Section IV,

$$P = \frac{\bar{\sigma}t}{R \left(1 - \frac{\nu}{2} \right)},$$

where

t = Wall thickness = 0.015 inch,
 R = Radius = 0.126 inch,
 ν = Poisson's ratio = 0.26, and
 $\bar{\sigma}$ = Effective stress.

FIGURE D-1. CALCULATED POWER PROFILE IN SPECIMEN C3C

FIGURE D-2. CALCULATED STEADY-STATE TEMPERATURE PROFILE IN SPECIMEN C3C AT END OF IRRADIATION

FIGURE D-3. FISSION GAS GENERATION IN SPECIMEN C3C

From the tensile test data at 1400°F, Figure 4-6, the measured yield strength of the cladding material (Type-347 stainless steel) was 23,700 psi. As seen from Figure D-4, this is somewhat higher than the published yield strength data for Type-347 stainless steel. Assuming the slope of the strength temperature relationship is the same for the cladding material as for the published data, then from the dashed line in Figure D-4 the yield strength at 1295°F is 26,000 psi. The effective stress at a strain of 0.91 percent from Figure 4-6 was 25,700 psi. Assuming that there is consistency between the yield strength and the effective stress for small temperature changes, the effective stress at a temperature of 1295°F can be found by the ratio

$$\begin{aligned}\bar{\sigma}_F &= \bar{\sigma}_{1400^\circ\text{F}} \left(\frac{\text{Y.S. } 1295^\circ\text{F}}{\text{Y.S. } 1400^\circ\text{F}} \right) \\ &= (25,700) \left(\frac{26,000}{23,700} \right) \\ &= 28,200 \text{ psi.}\end{aligned}$$

Solving for P:

$$P = \frac{(28,200)(0.015)}{(0.126) \left(1 - \frac{0.26}{2} \right)}$$

$$P = 3860 \text{ psig.}$$

The external cover-gas pressure was on the order of 440 psig.⁽¹⁰⁾ Therefore, the required internal pin pressure to cause a 1 percent cladding deformation should be approximately 3860 + 440 = 4300 psig.

This value is in the same order of magnitude as the calculated peak internal pressure generated during the transient (6,745 psig). The discrepancy between the two values could be a result of one or more of several factors, such as: an increase in cladding strength due to neutron bombardment (similar to cold working); leakage of a portion of the transiently-released gas from the fuel region to blanket or gas plenum; or failure of the pressure-transducer diaphragm, due to melting, before the time of peak pressure.

The most important fact to be noted, regardless of the apparent discrepancy, is that the post-transient analysis of both specimens C3B (see discussion for calculations) and C3C indicate that transiently-released fission gas could be a prime cladding failure mechanism in defining the transient limit of fast reactor ceramic fuel; and that the order-of-magnitude of the effect is calculable.

FIGURE D-4. YIELD STRENGTH VERSUS TEMPERATURE IN
TYPE 347 STAINLESS STEEL

APPENDIX E

CALCULATION OF FUEL SPECIMEN TEMPERATURES

Analytical methods employed to calculate fuel specimen temperatures were, with one exception, the same as in the previous work.⁽⁴⁾ In the earlier work, the TIGER-V heat transfer code was employed to estimate fuel and cladding temperatures. In this work, the Argus code⁽³¹⁾ was employed for these calculations for the reason that Argus is capable of varying material properties with temperature. In comparative cases, it was found that the TIGER-V and Argus results were essentially identical up to the point where melting of the fuel occurred. Once fuel melting was initiated, the TIGER-V computation would overestimate the cladding temperature since the code does not compensate for the rather large heat of fusion ($\sim 1180^{\circ}\text{F}$ equivalent) of the fuel. Thus, the higher calculated fuel temperatures resulted in higher calculated cladding temperatures. The geometrical model employed with the Argus code is illustrated in Figure E-1.

The material properties used in the input for Argus are presented in graphical form in Figures E-2 through E-9.

FIGURE E-1. GEOMETRIC MODEL FOR ARGUS CALCULATIONS

FIGURE E-2. THERMAL CONDUCTIVITY OF $\text{PuO}_2 - \text{UO}_2$ FUEL

FIGURE E-3. ρC_p OF MIXED OXIDE FUEL

FIGURE E-4. THERMAL CONDUCTIVITY FOR TYPE 347 STAINLESS STEEL (ANNEALED)

FIGURE E-5. ρC_p FOR TYPE 347 STAINLESS STEEL

FIGURE E-6. THERMAL CONDUCTIVITY OF NaK (78 PERCENT K, 22 PERCENT Na)

FIGURE E-7. ρC_p OF NaK

FIGURE E-8. THERMAL CONDUCTIVITY OF 6061 ALUMINUM

FIGURE E-9. ρC_p OF 6061 ALUMINUM

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Major contributions to the program were made by:

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Irradiation in GETR	R. D. Robertshaw
Post Irradiation Examinations	R. W. Darmitzel
Fuel Fabrication	E. F. Kurtz
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