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POST-IRRADIATION EVALUATION OF
A PLATE-TYPE UO_2 FUEL ELEMENT

JANUARY 1963

CONTRACT AT-11-1-GEN-14

BETTIS ATOMIC POWER LABORATORY, PITTSBURGH, PA.,
OPERATED FOR THE U. S. ATOMIC ENERGY COMMISSION
BY WESTINGHOUSE ELECTRIC CORPORATION

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H. B. Meieran

January 1963

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The premature failure of fuel Element M22, which had six compartments of 0.100-inch-thick, 96% TD $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuel, has been attributed to the large irradiation-induced solid volume swelling of the UO_2 fuels. This volume swelling was the result of incomplete homogenization during fabrication of the mixed and sintered U^{235}O_2 and U^{238}O_2 fuels in Element M22. In addition, heavy hydriding of the nickel-free Zircaloy-2 cladding occurred in the relatively hot areas adjacent to the fuel and to a lesser extent at the external cladding surfaces. By postulation, hydrogen was apparently formed by the radiolytic decomposition of water entrapped between fuel and cladding after formation of the initial cladding defect, and was absorbed by the cladding so rapidly that it could not diffuse adequately down the thermal gradient to the cold side of the cladding. The corrosion behavior of the cladding was as expected and did not contribute to the hydriding. Analysis of the CR-X-3 loop operating history indicates that no abnormal conditions external to Element M22 existed in the loop other than uranium from inpile intentionally defected fuel elements.

POST-IRRADIATION EVALUATION OF A PLATE-TYPE UO_2 FUEL ELEMENT

H. B. Meieran

I. INTRODUCTION

A. Purpose of Element M22

Element M22 was initially inserted into the X-3-m test in the CR-IV-X-3 loop in the NRX (Chalk River) reactor on May 6, 1960, to study the effects of bulk density, temperature, and quadrivalent metal ion additions on the irradiation behavior of bulk UO_2 in isostatic pressure-bonded plate elements. As information concerning the temperature dependency of plate element failures became available, this element was selectively moved into prescribed loop locations to maintain fuel temperatures below the temperature at which failure would be anticipated. In this manner the possibility of increasing specimen lifetimes by reducing sample operating heat fluxes was investigated.

B. Description of Element M22

Element M22 consisted of twelve nickel-free, Zircaloy-2 clad, 1.5-inch-long x 0.250-inch-wide x 0.100-inch-thick fuel compartments, six of which contained 96% TD bulk UO_2 fuel wafers (M22 tier 1-M22T1) and six of which contained 96% TD $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuel wafers (M22 tier 2-M22T2). Figure 1 shows fuel element geometry and location of all twelve compartments of Element M22. The fuel compositions are shown in Table 1 and the Zircaloy cladding ingot compositions are shown in Table 2. Element M22 was isostatic pressure bonded (IPB) at 1550°F for 4 hours at 10,000 psi in a bonding run arbitrarily called bonding run 1 at Battelle Memorial Institute. All fuel elements for the X-3-m test (inpile samples and controls) except Element M52 were bonded during this bonding run (M11*, M12, M21, M22*, M31*, M32, M41, M42*, M51, M61*, M62, M71, and M72*). Pre-irradiation corrosion and destructive evaluation data are shown in Table 3. The pre-irradiation dimensional, weight, and density data for Element M22 are included with the irradiation interim examination data shown in Appendix A and summarized in Table 4.

*Elements which were placed inpile. The fuel wafer and element fabrication procedures and all irradiation data were obtained from R. C. Daniel, Bettis Atomic Power Laboratory. Also, see Reference 1.

TABLE 1. MASS SPECTROMETRIC ISOTOPIC, CHEMICAL, AND EMISSION SPECTROGRAPHIC ANALYSES OF ELEMENT M22 FUEL

A. Mass Spectrometric Isotopic Analyses

Fuel Composition	Run No. (EMB)	w/o of Total Uranium Before Fabrication				w/o of Total Uranium After Fabrication				Radio-chem. Log No.
		U ²³⁴	U ²³⁵	U ²³⁶	U ²³⁸	U ²³⁴	U ²³⁵	U ²³⁶	U ²³⁸	
UO ₂ + 6 w/o ZrO ₂	72	0.160 ± 0.01	14.35 ± 0.06	0.05	85.48 ± 0.07	0.150 ± 0.01	14.53 ± 0.05	0.053 ± 0.008	85.26 ± 0.06	9371
UO ₂	111	0.158 ± 0.002	15.15 ± 0.06	0.063 ± 0.006	84.67 ± 0.04	0.160 ± 0.01	14.93 ± 0.05	0.054 ± 0.008	84.86 ± 0.06	9367

B. Chemical Analyses

Fuel Composition	Run No. (EMB)	Total U (%)	U (VI) (% of Total U)	H ₂ O (%)	Zr (%)	C (ppm)	Remarks
UO ₂ + 6 w/o ZrO ₂	72	87.7	5.4	0.275	--	--	Before addition of ZrO ₂ .
UO ₂ + 6 w/o ZrO ₂	72	83.1	5.1	—	4.34	330	After addition of ZrO ₂ .
UO ₂	111	87.8	6.2	0.17	--	210	--

C. Emission Spectrographic Analyses (ppm)

Fuel Composition	Run No. (EMB)	Al	Cr	Cu	Fe	Mg	Mn	Ni	Pb	Si	Sn	Zr
UO ₂ + 6 w/o ZrO ₂	72	83	<10	44	79	48	<10	157	11	167	<10	170*
UO ₂	111	9	<10	9	53	11	<10	14	<5	128	<5	50

*Before addition of ZrO₂.

TABLE 2. CHEMICAL ANALYSES AND MECHANICAL PROPERTIES OF
NICKEL-FREE ZIRCALOY-2 FROM INGOT MSM-28349

<u>Alloying Elements (w/o)</u>	<u>Top</u>	<u>Middle</u>	<u>Bottom</u>
Sn	1.46	1.43	1.41
Fe	0.132	0.128	0.130
Cr	0.115	0.110	0.1075
<u>Impurities (ppm)</u>			
Ni	20	20	20
Al	25	27	22
B	0.33	0.37	0.30
C	174	190	188
Cd	0.3	0.3	0.3
Cu	10	10	10
Hf	80	80	83
Mg	20	20	20
Mn	20	20	20
Mo	20	20	20
Pb	20	20	20
Si	38	38	25
Ti	20	20	20
U	--	--	--
V	20	20	20
W	20	20	20
H ₂	16	19	19
N ₂	44	40	40
O ₂	-----1450 to 1510-----		
<u>Mechanical Properties</u>			
Hardness, R _B	92.5-94	93-93.5	93-94
Tensile strength, psi	34,700	--	35,500
Yield strength, psi (2% Offset)	26,100	--	26,300
Elongation, %	40.5	--	41.0

C. Irradiation History of the Element

Element M22 was inserted into position 2 of the X-3 loop on May 6, 1960, with the identification end (Figure 1) oriented toward the bottom of the loop but above the maximum neutron flux axial position for the X-3-m test (Figure 2). The startup heat flux range for the six UO₂ compartments was 530,000 to 582,000 Btu/hr-ft², with an average of 560,000 Btu/hr-ft². The startup heat flux range for the six UO₂ + 6 w/o ZrO₂ compartments was 479,000 to 519,000 Btu/hr-ft², with an average of 497,000 Btu/hr-ft². The characteristics of the remainder of the samples listed in Figure 2 as well as the irradiation behavior thus far available are discussed in References 1 and 2.

NOTES:

1. EACH COMPARTMENT IS 0.250 IN. WIDE.
2. COMPARTMENTS ARE SEPARATED IN WIDTH DIRECTION BY 0.035-IN. STRIP.
3. END TABS ARE WELDED IN PLACE.
4. SURFACE NO. 1 AND EDGE NO. 1 ARE SHOWN, OPPOSITE SURFACE AND EDGE ARE NO. 2.

Figure 1. X-3-m Test Elements (12 Fuel Filler Design).

Figure 2. X-3-m Test Fuel Element Locations.

TABLE 3. PRE-IRRADIATION EVALUATION OF ELEMENT M22 INTEGRITY

Corrosion Test Data (6 days in 680°F, 2700 psi water; water conductivity: 0-2 μ mhos/cm; pH: 6.0-8.0)

<u>Weight Gain after Corrosion (mg)</u>		<u>Weight Change after Drying (mg)</u>	
<u>Specification</u>	<u>Observed</u>	<u>Specification</u>	<u>Observed</u>
10 - 30	18.6	<1	0.3

Destructive Testing Data (Destructive test evaluation of four compartments)

<u>Burst Pressure Range (psi)</u>	<u>Maximum Deflection over Rupture (mils)</u>	<u>Bond Quality*</u>	
		<u>Horizontal Bonds</u>	<u>Vertical Bonds</u>
4200 - 5200	29 - 55	A	A-5% B

*A - Bond not visible due to grain growth across it.

B - Bond visible due to precipitates at bond.

After 337.8 EFPD at an average burnup of 11.6×10^{20} fissions/cc for the UO_2 compartments and 10.3×10^{20} fissions/cc for the $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ compartments, the X-3-m test element axial loop loadings were revised to accommodate two irradiated J-16-1G ceramic seed elements, 15D and B72 (Reference 3), and to drop the heat flux and, hence, the fuel centerline temperatures of Element M22. At this time (July 24, 1961) Element M22 was placed in position 7 in the X-3 loop (below the maximum neutron flux position) with the identification end still oriented toward the bottom of the loop (Figure 2). Thus, the pair of compartments most highly depleted in U^{235} , 1A and 2A, became those with the lowest heat outputs in the element; the least depleted compartments, 1F and 2F, became those with the highest heat outputs.

After a total of 432.8 EFPD at an average burnup of 13.65×10^{20} fissions/cc in the UO_2 compartments and 12.15×10^{20} fissions/cc in the $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ compartments, Element M22 was repositioned to loop positions 4 and 5 in the X-3-n test. The X-3-m test ended November 20, 1961, as shown in Figure 3. Again, the identification end was oriented toward the bottom of the loop and the positions were below the maximum neutron flux positions.

Element M22 was continued under uninterrupted irradiation until 0430 hours on January 18, 1962, when the loop activity increased, indicating a failed fuel element; the failed element was subsequently identified as Element M22 when the X-3-n test was removed from the NRX reactor at 0201 hours on January 22, 1962. Thus, the element was operated for 93.5 hours after the initial defect was noted. The total exposure time for Element M22 at the time of its removal was 478.1 EFPD. It was found that UO_2 compartment 1E had swelled and leaked (as determined by a liquid nitrogen leak test). At the time of failure, Element M22 had a calculated UO_2 burnup range of 14.6 to 14.8×10^{20} fissions/cc (average of 14.75×10^{20} fissions/cc) and a $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ burnup range of 13.1 to 13.2×10^{20} fissions/cc (average of 13.15×10^{20} fissions/cc).

The heat flux at the end of life was 280,000 to 333,000 Btu/hr-ft² for the six UO_2 compartments and 249,000 to 296,000 Btu/hr-ft² for the six $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ compartments. The heat flux and burnup at the time of failure for the failed UO_2 compartment 1E were 320,000 Btu/hr-ft² and 14.75×10^{20} fissions/cc, respectively.

TABLE 4. SUMMARY OF INTERIM AND POST-IRRADIATION EXAMINATION DATA FOR ELEMENT M22

	<u>Inser-</u> <u>tion</u>	<u>Exam.</u> <u>No. 1</u>	<u>Exam.</u> <u>No. 2</u>	<u>Exam.</u> <u>No. 3</u>	<u>Exam.</u> <u>No. 4</u>	<u>Exam.</u> <u>No. 5</u>	<u>Exam.</u> <u>No. 6</u>	<u>Exam.</u> <u>No. 7</u>	<u>Exam.</u> <u>No. 8</u>
<u>Date</u>	5-6-60	7-26-60	9-19-60	11-21-60	12-19-60	1-23-61	2-27-61	4-19-61	6-26-61
<u>EFPD</u>	0	67.9	111.0	161.6	179.6	203.9	227.1	264.2	321.1
<u>Element:</u>									
Avg ΔW , mils	0	-2.4	-1.7	-1.2	-0.5	-0.3	-0.8	+0.2	-0.8
Range ΔW , mils	0	-3.1/-1.8	-2.4/-1.1	-2.5/+0.3	-1.5/+0.9	-1.3/+0.7	-1.8/+1.2	-1.3/+0.7	-1.9/+0.9
Max. ΔL , mils	0	--	--	--	--	--	+5.0	+4.0	+3.0
Δ Weight, mg	0	+10.6	+38.9	+54.4	+63.6	+70.7	+77.7	+81.0	+87.8
Weight gain, mg/dm ²	0	8.15	29.9	41.9	49.0	54.5	59.8	62.3	67.6
<u>M22T1⁽¹⁾</u>									
Avg fissions/cc x 10 ⁻²⁰	0	2.76	4.28	6.06	6.70	7.50	8.30	9.45	11.08
Avg Btu/hr-ft ² x 10 ⁻³	560	525	506	482	473	462	451	437	416
Avg Fuel $\bar{C}$ Temp, °F	2495	2370	2300	2215	2185	2145	2105	2055	1980
Avg Δt (mils) = % Δt	0	-0.15	+0.1	+1.5	1.6	1.7	2.1	2.6	4.2
Range Δt (mils) = % Δt	0	-0.5/+0.6	-0.5/+0.8	1.0/2.3	1.0/2.7	0.9/2.2	1.7/3.0	2.0/3.9	3.5/5.4
<u>M22T2⁽²⁾</u>									
Avg fissions/cc x 10 ⁻²⁰	0	2.41	3.71	5.48	6.03	6.59	7.32	8.44	9.93
Avg Btu/hr-ft ² x 10 ⁻³	497	466	450	427	420	413	403	389	370
Avg Fuel $\bar{C}$ Temp, °F	2270	2160	2100	2020	1995	1970	1935	1885	1815
Avg Δt (mils) = % Δt	0	+0.65	1.3	2.7	2.8	3.0	3.2	3.3	4.8
Range Δt (mils) = % Δt	0	0.5/1.1	0.9/1.8	2.2/3.1	2.3/3.2	2.8/3.0	3.1/3.5	3.0/3.6	4.7/4.9

TABLE 4 (Cont)

	Exam. No. 9	Exam. No. 9 ⁽³⁾	Exam. No. 10	Exam. No. 11	Exam. No. 12	Exam. No. 12 ⁽⁴⁾	Exam. No. 13	Bettis	Remarks
<u>Date</u>	7-24-61	7-24-61	9-18-61	10-23-61	11-21-61	11-21-61	1-22-62	2-9-62	
<u>EFPD</u>	337.8	337.8	381.8	409.2	432.8	432.8	478.1	478.1	
<u>Element:</u>									
Avg ΔW , mils	+0.2	--	+0.3	+0.6	-0.8	--	-0.5	+0.7	
Range ΔW , mils	-0.3/+1.1	--	+0.1/+0.7	+0.3/+0.8	-2.3/+0.8	--	-1.3/+0.5	+0.1/+1.3	
Max ΔL , mils	+3.0	--	+3.0	+5.0	+3.0	--	+8.0	+8.2	
Δ Weight, mg	+88.0	--	+95.3	+97.8	+101.8	--	+105.7	--	
Weight gain, mg/dm ²	67.8	--	73.3	75.3	78.3	--	81.4	--	
<u>M22T1⁽¹⁾:</u>									
Avg fissions/cc x 10 ⁻²⁰	11.60	11.60	12.68	13.2	13.65	13.65	14.75	14.75	
Avg Btu/hr-ft ² x 10 ⁻³	408	305	292	287	282	324	305	305	
Avg Fuel $\bar{C}$ Temp, °F	1950	1585	1540	1520	1505	1640	1575	1575	
Avg Δt (mils) = % Δt	4.6	--	5.4	6.0	5.9	--	9.9 ⁽⁵⁾	9.5 ⁽⁵⁾	Failed Comp. 1E: Δt = 17.6 mils at CR.
Range Δt (mils) = % Δt	4.1/6.0	--	4.6/6.3	5.1/6.7	5.3/6.8	--	8.0/13.6 ⁽⁵⁾	7.3/12.8 ⁽⁵⁾	Failed Comp. 1E: Δt = 17.7 mils at Bettis.
<u>M22T2⁽²⁾:</u>									
Avg fissions/cc x 10 ⁻²⁰	10.3	10.3	11.2	11.8	12.15	12.15	13.15	13.15	
Avg Btu/hr-ft ² x 10 ⁻³	364	267	256	252	248	287	271	271	
Avg Fuel $\bar{C}$ Temp, °F	1800	1450	1410	1400	1385	1505	1450	1450	
Avg Δt (mils) = % Δt	5.2	--	5.7	6.0	6.0	--	7.6	7.2	
Range Δt (mils) = % Δt	4.8/5.9	--	5.3/5.9	5.6/6.4	5.5/6.6	--	6.8/8.4	6.8/7.8	

(1) UO₂.(2) UO₂ + 6 w/o ZrO₂.

(3) M22 repositioned from position No. 2 to No. 7 at this time.

(4) M22 repositioned to positions No. 4 and 5 of the X-3-n test.

(5) Average or range of five non-failed UO₂ compartments.

Figure 3. NRX Vertical Flux Distribution

Thirteen interim examinations were conducted on Element M22 during its irradiation period at average time intervals of 2 months. The results of these interim examinations are listed in Table A-1, and a summary of the interim and post-irradiation examination data is given in Table 4. A complete analysis of the interim examination data and Element M22 operating characteristics is given in Section II.

The inpile operating history of this plate element was uneventful until its time of failure except for one incident noted during the seventh interim examination on April 19, 1961. At this time the bow measurement of Element M22 was 45 mils as compared with the pre-irradiation bow measurement of 18 mils. This increase in bow was attributed to the fact that Element M72 in adjacent position No. 3 had failed, presumably because of burnout caused by specimen distortion (Reference 1). The mode of failure was such that a section of fuel and cladding in the middle of Element M72 melted when the clad surface of the element came into contact with the holder assembly. The high thermal gradients induced in the holder caused the holder to warp slightly; thus, the supporting buttons holding Element M22 became compressed and caused the slight bowing in Element M22.

A complete analysis of the X-3 loop operating history of the X-3-m and X-3-n tests during the period of Element M22 irradiation is given in Appendix B.

II. EVALUATION OF PRE-IRRADIATION, IRRADIATION, AND POST-IRRADIATION DATA

The pre-irradiation analysis of Element M22 was obtained from Bettis files. The element was also given thirteen interim examinations at Chalk River and one series of post-irradiation examinations at Bettis (Tables 4 and A-1). The interim examinations at Chalk River consisted of visual observations, periodic immersions in liquid nitrogen to locate any failed compartments, and to determine length, width, thickness, weight, density, and bow measurements. The post-irradiation examination at Bettis consisted of visual observations, a liquid nitrogen leak test, and thickness, length, and width measurements (all nondestructive tests). All twelve compartments of the element were then punctured to determine the

TABLE 5. THICKNESS INCREASES AND FISSION GAS RELEASE MEASUREMENTS

Compartment No.	Fuel Composition	Unirradiated Density (% TD)	Calculated Burnup (fiss/cc x 10^{-20})	Thickness Increase Δt (mils) = % Δt	Corrected Gas Collecting Pressure (μ)	% Fission Gas Release
1A	100% UO_2	97.4	14.80	8.9	115.4	5.50
1B		97.4	14.79	7.6	99.6	4.64
1C		97.4	14.78	7.3	89.4	4.26
1D		97.4	14.76	12.8	192.6	9.20
1E		97.5	14.75	17.7	0	Leaked
1F		97.4	14.62	10.8	114.5	5.53
2A	$\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$	98.5	13.20	7.4	55.0	2.94
2B		97.5	13.17	6.8	49.0	2.62
2C		97.4	13.14	6.8	52.7	2.82
2D		98.1	13.11	6.7	37.0	1.99
2E		98.0	13.09	7.4	47.9	2.58
2F		98.6	13.04	7.8	54.5	2.94

extent of fission gas release from the fuel into the compartment void volume. (The results are shown in Table 5 where they are compared with the various initial fuel wafer densities and thickness increases of each fuel compartment.) The element was then sectioned with an abrasive cutoff wheel to furnish various samples for fuel burnup analysis, metallographic and X-ray examination of fuel and cladding, fuel density, cladding hydrogen analyses, and emission spectrographic analyses of the cladding. Figure 4 shows the sample locations in Element M22 and indicates their approximate sizes.

A. Visual Examination

During the periodic visual examinations of Element M22 at Chalk River, it was noted that only light crud deposits were building up on the surface of the element. Further examination of the fuel element at Bettis confirmed the fact that there was very little crud deposition on the surface of the element. This is shown in Figure 5 in a plan view of both surfaces of Element M22. Figure 6A shows a typical crud area on Element M22; this crud generally surrounds the outline of each compartment. The only visible corrosion product is that seen on the center of the cladding over most compartments. This product was very light and is shown in Figure 6B. Visual examination at magnifications up to 36X failed to locate the defective area.

B. Liquid Nitrogen Leak Test

The liquid nitrogen leak tests consisted of immersing the fuel element in a bath of liquid nitrogen for approximately 20 minutes so that the nitrogen could seep into any defective areas. The element was then placed in a bath of methyl alcohol. Any nitrogen gas entrapped in the defective areas bubbles off through the alcohol, leaving a trail of bubbles emanating from the defect area. In this manner, at both Chalk River and Bettis Laboratory, a small defect hole was found in the center of the fuel outer-rib edge in UO_2 compartment 1E on the identification surface of Element M22 (Figure 5).

Figure 4. Element M22 Sectioning (Schematic Diagram).

Figure 5. Surfaces of Element M22 Showing Light Crud Deposits after Irradiation; 60% Reduction, 1.5X.

(A) SURFACE OF ELEMENT M22 SHOWING TYPICAL CRUD DEPOSITS SURROUNDING FUEL COMPARTMENT 1E; 36X.

(B) SURFACE OF ELEMENT M22 SHOWING TYPICAL WHITE CORROSION PRODUCTS ON THE CLADDING OVER UO_2 COMPARTMENT 1E; 36X.

Figure 6. Crud and Corrosion Products on Element M22.

C. Thermal History

The thermal history for both the UO_2 tier (M22T1) and $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ tier (M22T2) of Element M22 is shown in Table 4 and is plotted as a function of burnup in Figure 7 (average for UO_2 compartments), Figure 8 (UO_2 compartment 1E), and Figure 9 (average for $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ compartments). The temperatures are calculated from parameters listed in Reference 1. Included in Figures 7 and 8 are the superimposed curves for the lower limit for the fuel centerline temperature failure in isostatic pressure-bonded UO_2 fuel elements (taken from Reference 1).

Figures 7 and 8 show the consequences of the loop position change of this element as described in Section I-C. As the element approached the lower limit of the temperature failure curve for these elements at 11.6×10^{20} fissions/cc, the average element temperature was reduced from 1950°F to 1585°F , corresponding to surface heat fluxes of $408,000$ and $305,000 \text{ Btu/hr-ft}^2$, respectively (Figure 7). Further element exposure to 13.65×10^{20} fissions/cc reduced the heat flux to $282,000 \text{ Btu/hr-ft}^2$ and the fuel centerline temperature to 1505°F . In order to raise the heat flux and yet stay below the limitations imposed by the gas-gap temperature controlled failures, the element was moved to a higher flux position so that its average surface heat flux was increased to $324,000 \text{ Btu/hr-ft}^2$ and its corresponding average fuel centerline temperature was increased to 1640°F . After continued exposure for 41.3 EFPD to an average burnup of 14.75×10^{20} fissions/cc,

compartment 1E of this element failed, and the element was removed after 45.3 EFPD .

The thermal history of individual compartment 1E is shown in Figure 8, where it may be seen that this fuel platelet operated at about the same heat flux value as the element average until the time that Element M22 was repositioned from X-3-m test loop position 2 to position 7 (at a burnup of 11.4×10^{20} fissions/cc). From this time until the end-of-life, compartment 1E operated at surface heat fluxes between 5 and 10% above the element average.

Although the heat flux and, hence, the fuel centerline temperatures of the UO_2 and $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuels were dropped after 337.8 EFPD and raised slightly at 432.8 EFPD , the fuel element did not

Figure 7. Thermal and Swelling History for Six UO_2 Fuel Compartments in Element M22 (M22T1).

Figure 8. Thermal and Swelling History for Failed UO_2 Compartment 1E in Element M22 (M22T1).

Figure 9. Thermal and Swelling History for Element M22 (M22T2)
(Average of Six $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ Fuel Compartments).

attain prior-to-failure burnup predicted from the previously established lower limit of the failure curve for both the average UO_2 fuel conditions and for the failed UO_2 compartment 1E (Figures 7 and 8).

It is interesting to note that an error of about 30% in the calculation of the surface heat flux of compartment 1E would be sufficient to account for failure, since the limiting temperature value for specimen failure would be exceeded. However, radiochemical and mass spectrometric burnup analyses, described in Section II-I, do not support this hypothesis. The existence of temporarily high local neutron fluxes or loop water flow stoppages also might be hypothesized, in which case Element M22 would have experienced a temporary overheating and temperature rise to the point of failure (2000°F at the burnup of Element M22); thus, the total burnup, as determined by mass spectrometric and radiochemical analyses, would not be significantly affected. However, the loop operating history, as described in Appendix B, does not support such a theory, and temperature excursions were specifically excluded as possible factors contributing to the failure of compartment 1E. Therefore, it must be concluded that the failure of Element M22 occurred at a lower temperature and burnup than anticipated from previous data obtained from samples in the X-1 and X-3 loop tests described in Reference 1.

Figure 9 shows the thermal history of Element M22 (M22T2), $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuel, which operated about 10% lower in surface heat flux than the UO_2 in Element M22 (M22T1) as a result of the difference in U^{235} loadings for these two fuels (Table 1).

D. Fuel Compartment Thickness Increase

Thickness measurements were taken at three locations on each compartment of Element M22 during all thirteen interim examinations and the final examination at Bettis (two at a point 1/4 inch in from the end of each fuel compartment and one in the center of each fuel compartment). The average and range of thickness (shown in Table 4 and plotted in Figures 7, 8, and 9) are those taken at the center of each compartment where the maximum swelling would be expected. Included in these figures are the curves taken from Reference 1. These curves define the fuel element swelling as a function of burnup for other bulk UO_2 fuel elements.

The swelling behavior of the UO_2 fuel (Figures 7 and 8) up to about 11×10^{20} fissions/cc is consistent with that observed in similar elements (also described in Reference 1), and is shown by the dotted lines representing a swelling rate of $0.32\% \Delta t / 10^{20}$ fissions/cc in the figures. However, at exposures greater than about 11×10^{20} fissions/cc up to failure, the rate of fuel swelling increased until failure occurred in compartment 1E at 14.7×10^{20} fissions/cc. This behavior has been attributed in Reference 1 to either closing up of the internal void volume such that the true swelling rate of $0.7\% \Delta V$ per 10^{20} fissions/cc is then operative, or to formation of a fission gas gap which caused a large temperature differential across the gap and which grows progressively larger with increasing exposure until failure occurs. This latter mechanism (Reference 1) is defined by the temperature-failure curves shown in Figures 7 and 8. Although this behavior at first glance is typical of the gas-gap type of failures, several differences between these data and those described in Reference 1 are apparent. First, the failure occurred at lower temperatures and exposures than would be anticipated. In this regard, it may be observed that the temperature-failure curves were established on specimens with U^{235} loadings and inpile operating conditions such that the "average" fuel temperature and "end-of-life" fuel temperature varied by no more than 20%. Therefore, if the "average" fuel temperature of 1920°F throughout specimen life for Element M22 is considered, then element failure would be anticipated by the failure curve. However, this hypothesis is not substantiated by other fuel samples whose lives were extended by lowering the temperature (described in Section III-A) or by the metallographic examination described in Section II-J. Another interesting difference in the swelling behavior of the UO_2 in Element M22 as compared with other plates which failed by gap formation is the apparent increase in the fuel swelling rate of all the fuel compartments at exposures above 11×10^{20} fissions/cc. In typical gas-gap failures, generally only the failed compartment shows this effect and the kinetics of the failure are quite rapid. Thus, the swelling behavior is more typical of solid volume swelling after closing of the internal porosity. Examination of the data presented in Figures 7 and 8 shows that the swelling rate of the UO_2 is $0.7\% \Delta V / 10^{20}$ fissions/cc above 11×10^{20} fissions/cc. This implies that the pores closed up at the rate of $\frac{\% \text{ open porosity } (= \% \Delta V)}{\text{burnup}} = \frac{2.5\%}{11 \times 10^{20} \text{ fissions/cc}} = \sim 0.22\% \Delta V \text{ per } 10^{20} \text{ fissions/cc}$, which is in reasonably good agreement with the value of $0.16\% \Delta V \text{ per } 10^{20} \text{ fissions/cc}$ indicated in Reference 1. Thus, these results indicate that the swelling behavior of the UO_2 fuel is consistent with previously reported results with the exception of the failed compartment 1E. The behavior of this compartment is discussed in greater detail in Section III-A.

E. Fuel Element Width and Length Changes

The fuel element width changes are shown in Table 4. The maximum increase of +1.3 mils (+0.160% ΔW) and the maximum change of -3.1 mils (-0.382% ΔW) can be classed as insignificant or within experimental error.

Figure 10. Weight Gain of Element M22 as a Function of Exposure Time.

The fuel element length changes are shown in Table 4. The maximum length increase observed was +8.2 mils (+0.0735% ΔL). This change is believed to be significant; since the length changes do not appear to be related to increasing element exposure, the change in length is attributed to differential thermal expansion of the fuel and cladding.

F. Weight Changes

The weight changes for Element M22 are given in Table 4 and are plotted as a function of exposure time (EFPD) in Figure 10. Superimposed on Figure 10 is the curve indicating the expected weight gain of those elements irradiated in the X-3 and X-1 loops. As shown in Figure 10 there is no abnormal weight gain for Element M22 as compared with other X-3 loop elements, although the weight gain itself is abnormally high. The rate of weight gain was discontinuous at interim examination 7; at this time intentionally defected Element X7N11 failed and was removed from the X-3 loop. After this time the X-3 loop contained no intentionally defected specimens. The rate of weight gain for Element M22 prior to the time of the failure of Element X7N11 was 0.24 mg/dm²/day; the rate of weight gain after the removal of Element X7N11 was 0.085 mg/dm²/day.

The high weight gain of Element M22 is consistent with that reported in Reference 1 for all of the X-3 loop specimens, and is attributed to the uranium which was leached out of the intentionally defected UO₂ fuel Element X7N11 by the loop process water. This uranium is presumed to precipitate on the surface of other elements, perhaps as a constituent of the crud deposit within the X-3 loop, and subsequently increases the corrosion rate of the Zircaloy cladding.

A corollary explanation for the excessive corrosion rate of X-3 loop elements may be maloperation of the pressure-bonding operation for all of the elements in the X-3-m test with the exception of Element M52. Element M52, with a corrosion rate much smaller than that of the other X-3-m elements bonded

in Batelle bonding run 1, was bonded in Batelle bonding run 2. This phenomenon is discussed further in Section III.

G. Fuel Element Density Changes

Element M22 was given only one interim density measurement; this was made during the twelfth examination (432.8 EFPD). (See Table A-1.) The volume change of the fuel element at this time was:

$$\begin{aligned}\Delta V &= \frac{\text{Twelfth exam. (weight of M22)}}{\text{(density of M22)}} - \frac{\text{Unirradiated (weight of M22)}}{\text{(density of M22)}} \\ \Delta V &= \frac{160.9808 \text{ g}}{7.7917 \text{ g/cc}} - \frac{160.8790 \text{ g}}{7.9200 \text{ g/cc}} = 20.7 - 20.3 \\ \Delta V &= 0.4 \text{ cc}\end{aligned}$$

It can be assumed that there was no significant weight or volume gain for Element M22 as a result of corrosion, and that all of the noted volume increase was caused by the swelling of the fuels. The initial volume of the fuels was:

$$\begin{aligned}V_{\text{fuel}} &= \frac{\text{weight of } \text{UO}_2}{\text{density of } \text{UO}_2} + \frac{\text{weight of } \text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2}{\text{density of } \text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2} \\ &= \frac{38.6420 \text{ g}}{10.6800 \text{ g/cc}} + \frac{37.1020 \text{ g}}{10.2500 \text{ g/cc}} \\ &= 3.62 + 3.63 \\ &= 7.24 \text{ cc}\end{aligned}$$

The percent volume increase was:

$$\begin{aligned}\% \Delta V &= \frac{\text{volume change of fuel in 12 compartments} \times 100\%}{\text{initial volume of fuel in 12 compartments}} \\ &= \frac{0.4}{7.24} \times 100\% \\ &= 5.5\%\end{aligned}$$

The average Δt for the twelve fuel compartments at the twelfth interim examination was 5.0%, Δt or 2.95% ΔV ; the average burnup at this time was 13.0×10^{20} fissions/cc. Therefore, the rate of volume increase as determined by the thickness measurements was 0.23% ΔV per 10^{20} fissions/cc, and the rate as determined by density measurements was 0.42% ΔV per 10^{20} fissions/cc. The initial rate of swelling reported in Reference 1 was 0.16% ΔV per 10^{20} fissions/cc. Therefore, the rates found for Element M22 were 44 to 160% greater than the rate given in Reference 1. The deviation of volume change from thickness change assumes that the volume change is one-half the thickness change. Since fuel compartments tend to swell into a "pillow" shape with approximately a paraboloid clad configuration, the actual volume change is down to two-thirds to three-fourths of the maximum fractional thickness changes. The discrepancy between the two measurements of volume change is at least partially explained by this correction.

H. Fission Gas Release

All twelve compartments of Element M22 were punctured with a 1/8-inch-diameter drill, and the evolved gases were collected. The results of the fission gas release measurements are shown in Table 5, where they are compared with the thickness increase for each compartment. Figures 11 and 12 show the fission gas release plotted as a function of burnup compared with the fission gas releases

obtained from other UO_2 (Reference 1) and $\text{UO}_2 + \text{ZrO}_2$ (Reference 2) fuel elements. The fission gas released from the UO_2 fuel (identified in Figure 11 as M22T1) is higher than the lower limit of gas release values reported in Reference 1. The fission gas release values for the six M22T2 $\text{UO}_2 + 6 \text{ w/o}$ ZrO_2 fuel compartments (Figure 12) are, on the other hand, generally equal to those obtained from other $\text{UO}_2 + \text{ZrO}_2$ fuel elements.

I. Burnup Analyses

Selected compartments from Element M22 were sectioned to obtain pieces of fuel to be analyzed for burnup of the U^{235} atoms (see Figure 4 for section locations). The fuel was dissolved from the cladding, mass spectrometrically analyzed for the U^{235} content, and radiochemically analyzed for the Cs^{137} content. The analysis for U^{235} gave the ratio of U^{235} in total U after irradiation and was compared with the pre-irradiation U^{235} /total ratio to determine the amount of U^{235} depleted. The Cs^{137} radiochemical analysis determined the amount of Cs^{137} present in the sample and, hence, the number of U^{235} fissions experienced by the fuel. The results of the burnup analysis are shown in Table 6 along with the calculated burnup values. It can be seen that both the mass isotopic and Cs^{137} burnup analyses are in good agreement with the values calculated from the neutron flux distribution and heat output of the X-3 loop. Therefore, the calculated burnup values are used throughout this report to evaluate the behavior of the fuel elements.

J. Metallography

All samples were examined in the as-polished, fuel-etched, and clad-etched conditions. The acid as-polished condition was that obtained after the sample had been polished on a polishing wheel with a Linde B abrasive and an acid solution of 25 cc of $\text{HNO}_3 + 25$ drops of HF in 450 cc of H_2O . The UO_2 fuel etchant was 99% H_2O_2 and 1% H_2SO_4 ; the etching time was nominally 25 seconds. The $\text{UO}_2 + 6 \text{ w/o}$ ZrO_2 fuel etchant was a 90-10% or 95-5% solution of H_2O_2 and H_2SO_4 ; the etching time was a maximum of 3 minutes. The clad etchant was a 60-40% solution of H_2O and HNO_3 mixed with 8 drops of HF.

1. External Corrosion Film

The "normal" corrosion film over the heat transfer surface of both the UO_2 and $\text{UO}_2 + 6 \text{ w/o}$ ZrO_2 compartments of Element M22 varied from 6 to 13 microns. Typical maximum thickness corrosion film micrographs are shown in Figure 13; average film thicknesses were observed to be approximately two-thirds of the maximum film thickness. Figure 13A shows the maximum corrosion thickness of 7μ for examined $\text{UO}_2 + 6 \text{ w/o}$ ZrO_2 compartments 2D, 2E, and 2F. Figure 13B shows the maximum corrosion film thickness of 6μ for non-failed UO_2 compartments 1D and 1F. Figure 13C shows the maximum corrosion film thickness of 12μ on the edge-seal weld (a non-heat-transfer surface). Figure 13D shows the maximum corrosion film thickness of 6μ over the failed UO_2 compartment 1E at a distance of approximately 400 mils from the defective area. Figure 13E shows the $8\text{-}\mu$ -thick corrosion film seen on the failed UO_2 compartment 1E approximately 300 mils from the defective area.

The metallographic examination of the corrosion film provided proof that the observed film thickness was in good agreement with the weight gain data shown in Table 4 and Figure 10. The average corrosion film thickness for Element M22 as determined above by micro-examination was 4.5μ (4.5×10^{-4} cm). Since a corrosion film of 1μ is equivalent to a weight gain of 15 mg/dm^2 (Reference 1), then a corrosion film thickness of 4.5μ is equivalent to a weight gain of 67.5 mg/dm^2 . The difference between

TABLE 6. BURNUP ANALYSES

Compartment No.	Fuel (g/cc)	Enriched U ²³⁵ (a/o)	Uranium (w/o)	Atoms U ²³⁵ /cc x 10 ⁻²¹	Radio-chem. Log No.	U ²³⁵ Analysis		Cs ¹³⁷ Analysis		Calculated* Burnup (Fiss/cc x 10 ⁻²⁰)
						a/o U Fiss	Fiss/cc x 10 ⁻²⁰	a/o U Fiss	Fiss/cc x 10 ⁻²⁰	
1A	10.68	14.93	87.8	3.59	9368	40.8	14.65	37.5	13.48	14.80
1B	10.67	14.93	87.8	3.59	None	--	--	--	--	14.79
1C	10.68	14.93	87.8	3.59	None	--	--	--	--	14.78
1D	10.68	14.93	87.8	3.59	None	--	--	--	--	14.76
1E	10.69	14.93	87.8	3.59	9369	39.8	14.30	39.8	14.30	14.75
1F	10.68	14.93	87.8	3.59	9370	40.1	14.42	37.8	13.58	14.62
2A	10.30	14.53	83.1	3.20	9372	41.1	13.2	44.9	14.39	13.20
2B	10.20	14.53	83.1	3.17	None	--	--	--	--	13.17
2C	10.19	14.53	83.1	3.16	None	--	--	--	--	13.14
2D	10.26	14.53	83.1	3.18	None	--	--	--	--	13.11
2E	10.25	14.53	83.1	3.18	9373	40.9	13.0	45.0	14.30	13.09
2F	10.31	14.53	83.1	3.20	9374	40.5	12.95	39.1	12.51	13.04

*Calculated from nuclear design curve parameters for CR-X-1 and CR-X-3 loops.

(A) MAXIMUM CORROSION FILM OVER A $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ FUEL COMPARTMENT; AS-POLISHED, 1000 X.

(B) MAXIMUM CORROSION FILM OVER A NON-FAILED UO_2 COMPARTMENT; AS-POLISHED, 1000 X.

(C) MAXIMUM CORROSION FILM ON AN EDGE WELD; AS-POLISHED, 1000 X.

(D) MAXIMUM CORROSION FILM OVER FAILED UO_2 COMPARTMENT 1E, 400 MILS FROM LEAK; AS-POLISHED, 1000 X.

(E) MAXIMUM CORROSION FILM OVER DEFECTIVE UO_2 COMPARTMENT 1E, 300 MILS FROM LEAK; AS-POLISHED, 500 X.

Figure 13. Typical External Corrosion Films on Element M22.

the weight gain of 81.4 mg/dm^2 determined from weight gain measurements and the weight gain of 67.5 mg/dm^2 determined by metallographic examination can be attributed to crud deposits on the surface of the element.

A localized, abnormally thick corrosion film was noted on the failed UO_2 compartment 1E in an area which eventually was found to contain the defective area around the cladding crack. The cladding crack was first observed to be 279 mils from the defective area (indicated by the liquid nitrogen leak test) and was examined for a total distance of 176 mils toward the defective area (Section II-J-5). This film, shown in Figure 14, reveals that the cladding crack eventually penetrated through the cladding where the heavy film existed. The thickness of the film was 26μ at a distance of 300 mils from the leak indicated by the liquid nitrogen leak test (Figure 14A), 16μ at 248 mils (Figure 14B), 28μ at 191 mils (Figure 14C), 10μ at 162 mils (Figure 14D), 16μ at 135 mils (Figure 14E), and 8μ at 108 mils from the leak (Figure 14F). This localized heavy corrosion film was not typical of other regions of the plate surfaces as described above and shown in Figure 13. Since this abnormally thick corrosion film was localized in the region of the cladding crack, it is presumed to be associated with the element failure.

2. Fuel-Clad Gap

Six sections from Element M22 were metallographically examined (see Figure 14). Met sample 1-2DT, a transverse section through the non-failed compartments 1D (UO_2) and 2D ($\text{UO}_2 + 6 \text{ w/o ZrO}_2$), is shown in Figure 15B. Met sample 1D-EL, a longitudinal section through UO_2 compartments 1D (non-failed) and 1E (failed), is shown in Figure 16. Met sample 2D-EL, a longitudinal section through the non-failed $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ compartments 2D and 2E, is shown in Figure 16C. Met sample 1-2ET, a transverse section through the failed UO_2 compartment 1E

(A) DEPTH IS 300 MILS; FILM THICKNESS IS 26μ ;
AS-POLISHED, 500X.

(D) DEPTH IS 162 MILS; FILM THICKNESS IS 10μ ;
ETCHED CLADDING, 250X.

(B) DEPTH IS 248 MILS; FILM THICKNESS IS 16μ ;
AS-POLISHED, 500X.

(E) DEPTH IS 135 MILS; FILM THICKNESS IS 16μ ;
ETCHED CLADDING, 250X.

(C) DEPTH IS 191 MILS; FILM THICKNESS IS 28μ ;
AS-POLISHED, 500X.

(F) DEPTH IS 108 MILS; FILM THICKNESS IS 8μ ;
AS-POLISHED, 250X.

Figure 14. Localized Heavy Corrosion Film over Failed UO_2 Compartment 1E. Depths Noted Are Distances from Leak Indicated by Liquid Nitrogen Leak Test.

(A) TRANSVERSE SECTION THROUGH NON-DEFECTIVE COMPARTMENTS 1F (LEFT) AND 2F (RIGHT); AS-POLISHED, 13X.

(B) TRANSVERSE SECTION THROUGH NON-DEFECTIVE COMPARTMENTS 1D (LEFT) AND 2D (RIGHT); ETCHED FUEL AND CLADDING, 13X.

Figure 15. Metallographic Samples from UO_2 Compartments 1D and 1F and from $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ Compartments 2D and 2F; 25% Photo Reduction.

(A) TRANSVERSE SECTION THROUGH FAILED COMPARTMENT 1E (LEFT) AND NON-FAILED COMPARTMENT 2E (RIGHT); ETCHED FUEL AND CLADDING, 13X.

(B) LONGITUDINAL SECTION THROUGH FAILED COMPARTMENT 1E (LEFT) AND NON-FAILED COMPARTMENT 1D (RIGHT); ETCHED FUEL AND CLADDING, 13X.

(C) LONGITUDINAL SECTION THROUGH NON-FAILED COMPARTMENTS 2E (LEFT) AND 2D (RIGHT); ETCHED FUEL AND CLADDING, 13X.

Figure 16. Metallographic Samples from UO_2 Compartments 1D and 1E and $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ Compartments 2D and 2E; 40% Photo Reduction.

approximately 400 mils from the defective area and non-failed $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ compartment 2E, is shown in Figure 16A. Met sample 1-ET, a transverse section through the failed UO_2 compartment 1E approximately 103 mils from the defective region, is shown in Figure 17. Met sample 1-2FT, a transverse section through the non-failed compartments 1F (UO_2) and 2F ($\text{UO}_2 + 6 \text{ w/o ZrO}_2$), is shown in Figure 15A.

The observed fuel-clad separation was generally small, and most of it may be attributed to the sectioning of Element M22 when the cladding stress was relieved. The magnitude of fuel-clad separation found in Element M22 is the same as that found in unirradiated elements and specifically in Element M21, the unirradiated control element for Element M22 (Figure 18).

The separation of fuel and cladding for the non-failed UO_2 compartments is shown in Figures 15, 16, and 20. The fuel-clad separation for the longitudinal section of UO_2 compartment 1D, Figures 16B and 20A, was 1.6 mils, and the separation in the transverse section of UO_2 compartment 1D was 0.8 mil,

Figure 17. Transverse Composite Section of Failed UO_2 Compartment 1E at the Cladding Crack Area. This Section Is Approximately 102 Mils from the Leak Indicated by the Liquid Nitrogen Leak Test; Etched Cladding, 13X.

Figures 15B and 20B. The transverse section was taken from the center of compartment 1D, and it can be expected to show the maximum fuel-clad separation of that compartment. The separation of 0.8 mil accounts for only 6.4% of the total swelling of 12.8 mils. The separation for the transverse section of UO_2 compartment 1F is negligible (Figures 15A and 20C). The one point of maximum "separation" in the center of this compartment was where one of the thermal cracks in the fuel contacted the inside clad surface. Apparently, then, the compartment swelling for the non-failed UO_2 compartments was primarily caused by solid volume swelling of the fuel.

The separation of the fuel and cladding of failed UO_2 compartment 1E is shown in Figures 16, 17, 21, and 25 to be a maximum of 1.9 mils. Figure 16B illustrates the longitudinal section located 500 mils from the center of compartment 1E. Figure 16A illustrates the transverse section located 190 mils from the center of compartment 1E. Figure 17 illustrates the transverse section on the other side of the center of compartment 1E, located 355 mils from the center. Figure 21 illustrates the transverse section located 528 mils from the center of compartment 1E. Figure 25A illustrates the transverse section located 549 mils from the center of compartment 1E. The separation of 1.9 mils accounts for 10.7% of the total compartment swelling of 17.7 mils and is, therefore, indicative of the beginning of a fuel-clad separation and a fission gas gap.

Figure 15A illustrates the transverse section taken from the center of compartment 2F. Figure 15D illustrates the transverse section taken from the center of compartment 2D. Figure 16C illustrates the longitudinal sections taken from the ends of compartments 2D. Figure 16C illustrates the longitudinal sections taken from the ends of compartments 2D and 2E. Figures 16A and 32F illustrate the transverse section taken from the center of compartment 2E.

The separation of the fuel and cladding for the $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ compartments is shown in Figures 15, 16, and 32 to be about 0.4 mil and amounts to less than 5.1% of the total maximum swelling (7.8 mils) encountered in any $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ fuel compartment.

In the autocatalytic gas-gap failure mechanism characteristic of plate oxide fuel elements the temperature-failure curve, shown in Figures 7 and 8, is exceeded, fission gas is released which deflects

(A) VIEW SHOWING FUEL-CLAD INTERACTION ON $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ FUEL COMPARTMENT; ETCHED CLADDING, 250X.

(B) VIEW OF A UO_2 FUEL COMPARTMENT SHOWING NO FUEL-CLAD INTERACTION; ETCHED CLADDING, 250X.

(C) VIEW OF A UO_2 FUEL COMPARTMENT SHOWING FUEL-CLAD INTERACTION; ETCHED CLADDING, 250X.

Figure 18. Fuel-Clad Interaction in Unirradiated Control Element M21.

the cladding and elevates the fuel temperature, and causes still greater gas release and further deflection of the cladding. It is concluded, from the foregoing observations and from the fact that Element M22 was operated under conditions less severe than those causing gas-gap failure, that the gas-gap mechanism did not contribute significantly to the failure of Element M22. The compartment swelling is attributed primarily to the solid volume swelling of the fuel.

3. UO₂ Fuel Structures

A typical unirradiated as-polished and etched UO₂ fuel structure from control Element M21 (M21T1) is shown in Figures 19A and 19B. It is seen here that the typical grain size is approximately 15 μ . The porosity regions are voids formed during the fuel wafer fabrication. This porosity, which generally closed up during irradiation, is illustrated in both the failed and non-failed UO₂ microstructures of Figures 20, 21, 22, and 23. The new areas of porosity, which are results of fission gas bubbles, appear in two types of locations: (1) as a general distribution of small bubbles throughout the UO₂ matrix and (2) as clumps of bubbles located throughout the matrix. The general distribution of bubbles seen in Element M22T1 (Figure 21) is normal for UO₂ fuel at a burnup of less than 20×10^{20} fissions/cc in which an elevated temperature has been reached due to a gas gap (Reference 1). However, the clumps of bubbles, which are evident as "pockets" of bubbles in the as-polished fuel microstructures (Figures 22 and 23) or "stained" regions in the fuel-etched microstructures, encompass between 5 and 10% of the total surface area viewed in Element M22 (M22T1). This clumping of bubbles is not typical of microstructures observed in UO₂ or UO₂-based fuel materials, but is similar to the structures observed in enriched UO₂ particles which have been dispersed in matrices such as those of ZrO₂ (Reference 2), BeO, and Al₂O₃. It is evident from these results that inhomogeneity existed in this fuel material to the extent that clumps of enriched UO₂ existed in the fuel.

During the fabrication of Element M22 and other similar elements, quantities of powders (<1 μ in diameter) of U^NO₂ and fully enriched U^EO₂ are mixed to give the proper U²³⁵ enrichment required to furnish the designed heat flux. In Element M22T1 this ratio was approximately 15% U^EO₂ and 85% U^NO₂. During the subsequent sintering operations, portions of the U^EO₂ powder apparently agglomerated and were not homogenized with the bulk of the U^NO₂. Assuming that the "stained" areas and regions of clumped fission gas bubbles once contained fully enriched UO₂, then 5 to 10% of the total fuel matrix consisted of clumped highly depleted U^EO₂ particles. A typical unetched region for both failed and non-failed compartments is shown in Figures 23A and 23B. Therefore, one-third to two-thirds of the initial U^EO₂ content was not fully homogenized with the rest of the UO₂. Since the ratio of U²³⁵ atom density in 5 to 10% of the fuel to that in the whole matrix was 93% (fully enriched uranium) to 15% (enrichment of the total matrix), the localized burnups were in excess of 90×10^{20} fissions/cc ($93/15 \times 14.7 \times 10^{20}$ fissions/cc) in the UO₂ fuels in both the failed and non-failed compartments. Correspondingly, if two-thirds of the U^EO₂ were inhomogeneously distributed, the fission depletion of the matrix was less than 6.5×10^{20} fissions/cc.

There seemed to be very little difference between the fuel structures in the failed and non-failed UO₂ compartments, although general porosity in the failed compartment 1E (Figure 21) was higher than that in the non-failed compartments (Figure 20). The greater degree of general porosity seen in the center of failed compartment 1E was probably due to the presence of steam or water, as previously noted (Reference 1). The UO₂ grain size was, in general, only slightly (if at all) smaller than that found in unirradiated material in all of the fuel compartments in Element M22 (M22T1). In addition,

(A) TYPICAL UNIRRADIATED UO_2 FUEL
IN ELEMENT M21(M2IT1);
AS-POLISHED, 400X.

(B) TYPICAL UNIRRADIATED UO_2 FUEL
IN ELEMENT M21(M2IT1);
ETCHED, 400X.

(C) TYPICAL UNIRRADIATED $\text{UO}_2 + 6 \text{ w/o}$
 ZrO_2 FUEL IN ELEMENT M21(M2IT2);
AS-POLISHED, 400X.

(D) TYPICAL UNIRRADIATED $\text{UO}_2 + 6 \text{ w/o}$
 ZrO_2 FUEL IN ELEMENT M21(M2IT2);
ETCHED, 400X.

Figure 19. Unirradiated Fuels in Control Element M21.

(A) LONGITUDINAL COMPOSITE OF UO_2 COMPARTMENT 1D; AS-POLISHED, 75X.

(B) TRANSVERSE COMPOSITE OF UO_2 COMPARTMENT 1D; AS-POLISHED, 75X.

(C) TRANSVERSE COMPOSITE OF UO_2 COMPARTMENT 1F; AS-POLISHED, 75X.

Figure 20. Composite Sections through Non-failed UO_2 Compartments in Element M22; 55% Photo Reduction.

Figure 21. Transverse Composite of Fuel in the Center of Failed UO_2 Compartment 1E; 50% Photo Reduction, As-polished, 100X.

the grains were all delineated by the preferential attack of the UO_2 etchant upon the fission products located at the grain boundaries (Reference 1). Figure 22 shows the fuel structure resolved in a longitudinal section taken from the failed UO_2 compartment; this is typical of all fuels in Element M22T1. Figure 23 illustrates the grain size inhomogeneities existing in the fuel. In Figure 23 coarse grain size is noted at the fuel surface in a non-failed UO_2 compartment. This behavior is believed to be another indication of the original micro-inhomogeneity of the fuel material.

Several white precipitates were found in both the failed and non-failed fuels (Figure 23B) in regions of the unetched fuel structures. The fact that the precipitates were found in the highly depleted regions is indicative of the localized high fission depletions. These precipitates are presumed to be compounds or solid solutions of fission products and precipitated uranium (Reference 1).

An important observation made from Figures 22 and 23 is the existence of UO_2 grain boundary cracks which emanate from the localized high burnup regions. Such microcracks are unusually high in depleted UO_2 because of its irradiation induced plasticity (Reference 1). If the localized regions of high $\text{U}^{\text{E}}\text{O}_2$ content were irradiated to 90×10^{20} fissions/cc, these regions would expand about 63% in volume.* Apparently, the cracking of the UO_2 matrix by the enriched UO_2 particle expansion coupled with the low plasticity of the matrix resulting from its low depletion (6.5×10^{20} fissions/cc) caused both the high gas release shown in Figure 11 and the increased swelling shown in Figures 7 and 8.

In the $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ fuels, prepared by a double sintering process in which a sintered wafer is ground and mixed between the two sintering operations and where homogeneity is much greater than for the singly sintered UO_2 samples, no localized regions of high deflection similar to those shown in Figures 22 and 23 are visible.

4. Hydride Formation on the Cladding in Failed UO_2 Compartment 1E

Examination of the failed UO_2 compartment 1E revealed that extensive internal and external zirconium hydride precipitation occurred on the Zircaloy cladding and rib components. This phenomenon was observed in all three metallographic samples of the compartment viewed in both the transverse and longitudinal directions. The presence of the zirconium hydride along the internal cladding surface was

* $\frac{0.7\% \Delta V}{10^{20} \text{ fissions/cc}} \times 90 \times 10^{20} \text{ fissions/cc} = 63\% \Delta V$

(A) FUEL AT FUEL-CLAD INTERFACE;
AS-POLISHED, 400X.

(B) FUEL AT FUEL COMPARTMENT
CENTER; AS-POLISHED, 400 X.

(C) FUEL AT FUEL-CLAD INTERFACE; AS-POLISHED, 400X.

(D) FUEL AT FUEL-CLAD INTERFACE;
ETCHED, 400X.

(E) FUEL AT FUEL COMPARTMENT
CENTER; ETCHED, 400X.

(F) FUEL AT FUEL-CLAD INTERFACE; ETCHED, 400X.

Figure 22. Longitudinal Sections of Fuel from Failed UO_2 Compartment 1E; 40% Photo Reduction.

(A) ETCHED CLADDING; 250X.

(B) SECTION OF ETCHED FUEL SHOWING
UNETCHED AREA; DIAMETER OF
UNETCHED MATERIAL IS 45μ ; 400X.

(C) AS-POLISHED FUEL; 400X.

Figure 23. Longitudinal Fuel and Cladding Microstructures of a Fuel-Rib-Clad Corner
in a Typical Non-failed UO_2 Compartment; 20% Photo Reduction.

Figure 24. Typical Composite of Hydrogen Gradient in Cladding; Etched, 250X.

an observation not previously noted, since hydrogen is known to diffuse from the hot zones of the cladding to the colder zones (Reference 3). A general observation of the pattern of hydriding was that for all regions of solid internal hydride (estimated as 16,000 ppm of H_2) there were parallel regions of excessive hydride (5000 to 10,000 ppm of H_2) present on the external cladding surface. Figure 24 shows a typical composite of the internal and external hydrided cladding surfaces observed on all viewing planes. Cracks running parallel to the cladding surface are visible in the brittle internally hydrided region. Several planes were viewed in all three metallographic samples to examine the geometrical shape of the hydriding and estimate its contour within the regions of compartment 1E. Figures 16, 17, and 25 through 28 are selected micrographs showing the extent of the hydriding in all viewing planes. Figures 29 and 30, respectively, exhibit the two-dimensional analysis and three-dimensional estimate of the hydride contour within compartment 1E.

(A) COMPOSITE OF FUEL-CLAD INTERFACE; AS-POLISHED, 75X.

(B) COMPOSITE OF CLADDING; ETCHED, 100X.

(C) NON-HYDRIDED FUEL-RIB-CLAD
CORNER; ETCHED, 100X.

(D) HYCRIDED FUEL-RIB-CLAD
CORNER; ETCHED, 100X.

Figure 25. Hydride Structures in Cladding of Failed UO_2 Compartment 1E. Distance of Transverse Plane from Leak Indicated by Liquid Nitrogen Leak Test Is 300 Mils. Photo Reduction Is 70%.

(A)

(B)

(C)

(D)

Figure 26. Hydride Structures in Cladding of Failed UO_2 Compartment 1E. Depth of Section Plane from Leak Is 103 Mils. All with Etched Cladding, 100X.

(A)

(B)

COMPARTMENT 1E

COMPARTMENT 2E

(C) CORNER CONTAINING
CLADDING CRACK

(D)

Figure 27. Transverse Section of Failed UO_2 Compartment 1E Showing Extent of Hydride at Fuel-Rib-Clad Corners. This Section Is 420 Mils from the Leak Indicated by the Liquid Nitrogen Leak Test. Part of Compartment 2E Is Shown at Far Right. Photo Reduction 55%, Etched Cladding, 100X.

(A) SECTION OF CLADDING SIDE WHICH DID NOT HAVE A CLADDING CRACK; ETCHED CLADDING, 75X.

(B) SECTION OF CLADDING SIDE CONTAINING A CLADDING CRACK ETCHED CLADDING, 75X

Figure 28. Typical Longitudinal Section of Failed UO_2 Compartment 1E Showing the Extent of Hydride Formation in the Cladding. Distance from the Leak Indicated by the Liquid Nitrogen Leak Test Is 733 Mils to the Left of Sections Shown. Photo Reduction 30%, Etched Cladding, 75X.

Figure 29. Two-Dimensional Hydride Pattern on Cladding over UO_2 Compartment 1E, Element M22.

Figures 17, 25, and 26 show the hydride found in the transverse metallographic sample which contains the cladding crack (discussed later in this section). The sample was viewed at several planes to follow the patterns of the hydride and the cladding crack. The viewing planes ranged from 103 to 300 mils from the area which contained the compartment failure indicated by the liquid nitrogen leak test. Figure 25 shows the planar hydride observations at a distance of 300 mils from the leak; Figures 17 and 26 show the planar observations at a depth of 103 mils. The hydride, both internal and external, was present in three of the four fuel-rib-clad corners and varied in width from 5 to 60 mils and in depth from 1 to 5 mils. In several viewing planes at depths of 300 mils (Figure 25B) and 173 mils the regions of hydride extended over the internal clad surface at the center of the compartment. This phenomenon was the exception rather than the rule and indicated that the planar spread of the hydrided regions was irregular. In one corner no hydride was visible at all in the region containing the cladding crack (Figure 26C). This observation, attributed to the fact that the hydrogen generated internally was able to escape from this region through the cladding crack, is believed to be pertinent and is discussed in greater detail in Section II-J-5 and Section III.

The internal and external hydride precipitates were also evident in transverse and longitudinal metallographic samples taken from the other end of the failed compartment 1E at distances greater than 400 mils from the defective area. Figure 27 shows the hydride formations in all four corners of a transverse section taken through the failed compartment at a distance of 400 mils from the leak. This figure illustrates the typical hydride structure as seen on two viewing planes in the sample (vertical plane separation of 20 mils). The hydride present on the cladding of all four fuel-rib-clad corners is approximately 6 mils thick and 25 to 65 mils wide. Figure 27C, a view of the fuel-rib-clad corner with

Figure 30. Three-Dimensional Hydride Contour in Cladding over UO_2 Compartment 1E, Element M22.

the cladding crack shows the localized large Zircaloy grains formed during the fabrication of Element M22 which are a characteristic of the pressure bonding treatment. Figure 16A shows a general transverse view of the whole compartment with the relative distances and depths of the hydride structure quite evident. Apparent in Figures 16A, 16C, and 27C is the absence of the hydride layer in adjacent non-failed compartments.

Figure 28 shows the internal and external hydride in a longitudinal metallographic sample taken near the end of the failed UO_2 compartment at a depth of 733 mils from the failed area. This figure illustrates the typical hydride structure as seen on two viewing planes in this sample (vertical plane separation of 20 mils). Figure 16B shows a general view of this longitudinal section from compartment 1E. It can be seen in these figures that the hydride begins at a point $1/8$ inch away from the center rib between failed UO_2 compartment 1E and non-failed UO_2 compartment 1D. The zone of solid internal hydride is about 4.8 mils thick and extends for a distance of 92 mils on one clad surface and for a distance of 88 mils on the clad surface with the cladding crack.

From the examination of the micrographs, it was possible to plot a two-dimensional hydride pattern present at the internal cladding surfaces over both sides of compartment 1E (Figure 29). From this pattern a three-dimensional estimate of hydride contour through the compartment was drawn (Figure 30).

TABLE 7. ESTIMATE OF Zr_2H_3 AND H_2 CONTENT IN CLADDING OVER COMPARTMENT 1E

Semi-Elliptical Rod Corner No.	Average Semi-Major Axis, a (in.)	Average Semi-Minor Axis, b (in.)	Length (in.)	Surface Area*		Volume **	
				in. ²	cm ²	in. ³ x 10 ⁴	cm ³ x 10 ³
1	0.0188	0.005	0.5	0.019	0.123	0.74	1.21
2	0.015	0.005	1.0	0.030	0.193	1.18	1.93
3	0.025	0.005	1.0	0.050	0.323	1.97	3.23
4	0.025	0.005	1.0	0.050	0.323	1.97	3.23
				Equivalent Length (in.)			
Semicircular End No.							
1	0.0750	0.005	0.628	0.094	0.606	3.75	6.05
2	0.0250	0.004	0.470	0.024	0.151	0.74	1.21
3	0.0375	0.005	0.505	0.038	0.245	1.19	1.95
4	0.0375	0.005	0.505	<u>0.038</u>	<u>0.245</u>	<u>1.19</u>	<u>1.95</u>
				0.343	2.209	12.73	20.8

Total Weight of Zr_2H_3 ($\rho = 5.6 \text{ g/cm}^3$) = 116 mg.

Total Weight of H_2 (16,000 ppm H_2 in Zr_2H_3) = 1.9 mg.

*Surface area = 2 x average semi-major axis x length = 2 x a x l.

**Volume = $\frac{\pi a b l}{2}$.

†Equivalent length = πR (semicircular end)

where $R = \frac{0.250 + (2 a)}{2}$ = 0.200 for end No. 1
 = 0.150 for end No. 2
 = 0.161 for end No. 3
 = 0.161 for end No. 4

From Figures 29 and 30 the surface area, volume, and weight of the internal solid Zircaloy hydride can be estimated. The hydride contour is generally composed of eight sections and two major parts, semi-elliptical rods of hydride on the four corners and semi-elliptical semi-circular rings of hydride. The estimate of the volume and weight of hydride for each of the eight sections is shown in Table 7. The total estimated surface area of hydride adjacent to the fuel is 2.21 cm²; the total estimated volume of hydride is 0.0208 cm³, and the weight is 116 mg of Zr_2H_3 or 1.9 mg of H_2 .

5. Cladding Crack

During the examination of the metallographic sample taken from the failed UO_2 compartment 1E near the defective area (Met sample 1-ET), a crack was found extending through the clad near the outer rib. The crack was first found at a distance of 279 mils from the defective area (see previous subsection 4) and was examined for a total distance of 176 mils toward the defective area before examination of the sample was completed, although the crack was still present. Figure 26 shows the typical structure of the crack which appears to start approximately 1/16 inch from the fuel-rib-clad corner where the maximum compartment stress is normally concentrated. Examination of a fuel compartment from unirradiated control

(A) NOTE CRACKS THROUGH CLADDING AND SLIGHT FUEL-CLAD INTERACTION AT COMPARTMENT CORNER; ETCHED, 150X.

(B) SECTION SHOWN IN (A), BUT ILLUMINATED WITH POLARIZED LIGHT; ETCHED CLADDING, 150 X.

Figure 31. Fuel-Rib-Clad Corner of Failed Pressure-Burst-Tested Fuel Compartment in Unirradiated Control Element M21.

not excessively unless the grains are considerably larger than sizes ASTM 4 to 5 (6 to 12 and 12 to 24 grains/in.², Reference 4).

The cladding in the general vicinity of the crack did not exhibit any hydride structures. This is interpreted to indicate that generated hydrogen which was trapped and absorbed near the other three fuel-rib-clad corners escaped through the crack to the coolant at the fourth corner.

Element M21 indicates that there was some fuel-clad reaction at the fuel-rib-clad corners, which was also the point of failure in a pressure burst test (failure occurred at 1200 to 5200 psi, Table 3). This failed corner is shown in Figure 31.

The crack in the failed compartment shows several peculiarities. First, the crack did not occur at the fuel-rib corner where maximum stress occurs and where almost all cladding failures due to internal pressure initiate both in irradiated and unirradiated specimens. Second, the cracks in as-bonded Zircaloy (bonded in the $\alpha + \beta$ range at 1550°F) usually follow the grain-boundaries (Figure 31) as a result of the precipitation of embrittling intermetallic compounds at the grain-boundaries during fabrication. However, in this sample the crack was transgranular (Figure 26) and showed a well-defined cleavage path through each grain. Finally, the crack penetrated the cladding through a region of coarse grain-size often found at this location during pressure-bonding because of critical strain. With the coarse grain size observed here, cleavage failure is preferred to grain-boundary rupture in irradiated Zircaloy-2.

These observations are not pertinent to fuel elements in which the beta heat treatment applied to the as-bonded elements eliminates all trace of the initial alpha grain size as well as of the grain boundary precipitate. The heavy corrosion film shown in Figure 14 in the region of the crack may also result from unusual corrosion attack on the coarse-grained Zircaloy (minimum of 10 grains/in.² at 100X or grain sizes of ASTM 4) at this location. Similar coarse grains at the corners of fuel compartments have been noted in other elements. They reduce cladding ductility locally but

6. $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ Fuel Structures

A typical unirradiated, as-polished and etched $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuel structure from control Element M21 (M21T2) is shown in Figures 19C and 19D, where the typical grain size is approximately 90μ . This original grain size around the fuel-clad interfaces was preserved during irradiation (Figures 32 and 33), although the general fuel structure could not be etched by standard metallographic techniques. The unresolved structure at the center of the wafers is believed to result from precipitation of the ZrO_2 -rich tetragonal phase at the higher temperatures obtained toward the center of the wafer. The initiation of precipitation at the grain boundaries of the supersaturated UO_2 -base solid solution is clearly evident in Figure 32B. This precipitation pattern is quite evident in the micrographs of compartments 2D and 2E shown in Figures 16A and 16C.

Figures 32D, 32E, 33A, and 33B show the fuel structure at the fuel-rib-clad corners which is similar to that found in the center of the compartments at the fuel-clad interfaces. There are regions in the fuel, both at the fuel-rib-clad corners and near the fuel-clad interfaces, which contain areas of unidentified fuel structures (Figure 33C). These structures probably result from inclusions or voids in the original fuel wafers.

7. Gray Phase

The gray phase (Reference 1) was present in all six of the examined fuel compartments to a depth of about 7.5μ (Figures 20, 23, 32, and 33), appearing as a continuous film in the UO_2 fuel compartments and discontinuous in the $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuel compartments. This gray phase, which has the same appearance and texture as the external ZrO_2 corrosion film, was interpreted to be the result of the oxidation of the internal cladding surface by the oxygen freed from the UO_2 fuel lattice upon fissioning of the uranium atoms.

The gray phase has been found at all fuel-cladding interfaces for UO_2 fuel elements irradiated to greater than 6×10^{20} fissions/cc. The extent of the gray phase is a function of the extent of burnup, the exposure time, and the amount of fuel-cladding surface contact. In Element 2H1, which failed after 141 EFPD and at a burnup of 17×10^{20} fissions/cc (Reference 1), the gray phase was very discontinuous. However, in Element 4L1, containing low density fuel (Reference 1) where the fuel-cladding contact was greater than that for Element 2H1 because of the Zircaloy fingers extruding into regions of open porosity, the gray phase was thick and continuous after 139 EFPD with a burnup of 17×10^{20} fissions/cc. The gray phase on Element M22T1 (UO_2 fuels), shown in Figures 22 and 23, is similar in extent to that observed in Element 4L1. The continuity of this phase in this sample may also be an indication of the local fission depletion inhomogeneities previously discussed.

The gray phase in Element M22T2 ($\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuels) was not as continuous as in Element M22T1 (Figures 32 and 33). It covered approximately 50% of the entire fuel-clad interface to a depth of 7.5μ . This is equivalent to a volume of 25.3×10^{-3} cc, or 14.2×10^{-3} grams of ZrO_2 (0.70×10^{20} moles of O_2). The fuel of Element M22T2 experienced 13.2×10^{20} fissions/cc, releasing 8.09×10^{20} molecules of O_2 within any specific compartment for each O_2 molecule released per fission. Since 0.7×10^{20} molecules of oxygen are required to form the observed layer, only 9% of the total number of molecules hypothetically released could be accounted for as reacting with the cladding. This figure may be compared with that of 18 to 25% found in other urania-zirconia fuels (Reference 2).

(A) TYPICAL FUEL-CLAD INTERFACE COMPOSITE FOR ALL COMPARTMENTS CONTAINING $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ FUELS; ETCHED FUEL, 75X.

(B) FUEL AT FUEL-CLAD INTERFACE; ETCHED FUEL, 400X.

(C) FUEL AT CENTER OF COMPARTMENT; ETCHED FUEL, 400X.

(D) TYPICAL FUEL AT FUEL-RIB-CLAD CORNER, ADJACENT TO FAILED UO_2 COMPARTMENT 1E; ETCHED FUEL, 400X.

(E) SAME AS (D) EXCEPT AS-POLISHED.

(F) TYPICAL FUEL-CLAD COMPOSITE FOR ALL COMPARTMENTS CONTAINING $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$; AS-POLISHED, 75X.

Figure 32. Transverse Section of $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ Fuel Compartment 1E; 55% Photo Reduction.

(A) TYPICAL $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ FUEL AT FUEL-RIB-CLAD CORNER SHOWING GRAY PHASE AND ONE OF SEVERAL ZIRCALOY FINGERS EXTENDING INTO OPEN PORE; AS-POLISHED, 400X.

(B) SAME AS (A) EXCEPT FUEL ETCHED.

(C) UNETCHED REGION OF ETCHED FUEL NEAR FUEL-CLAD INTERFACE OF LONGITUDINAL SAMPLE FROM COMPARTMENT 2D; 400X.

Figure 33. Fuel Structures in $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ Fuels from Element M22 (M22T2); 25% Photo Reduction.

TABLE 8. X-RAY DIFFRACTOGRAM ANALYSES

Nominal Fuel Composition	Element No.	Compartment No.	Burnup (fiss/cc x 10 ⁻²⁰)	Cell Size, A _o (Å)	Equivalent ZrO ₂ Composition*	
					(m/o ±1.4)	(w/o ±0.6)
UO ₂ + 6 w/o ZrO ₂	BHT	Control	0	5.421 ±0.005	13.5	6.72
UO ₂ + 6 w/o ZrO ₂	M21T2	Comp. 2A	0	5.430 ±0.005	10.95	5.30
UO ₂ + 6 w/o ZrO ₂	M22T2	2D	13.2	5.440 ±0.005	8.15	3.90
UO ₂ + 6 w/o ZrO ₂	BHT	B	21.4	5.423 ±0.005	12.9	6.35
100% UO ₂	M22T1	1D	14.75	5.469 ±0.005	--	--

*Calculated according to Vegard's Law:

$$\begin{aligned}
 \text{Mole fraction ZrO}_2 &= \frac{\text{cell size of sample} - \text{cell size of pure ZrO}_2}{\text{cell size of pure UO}_2 - \text{cell size of pure ZrO}_2} \\
 &= \frac{\text{cell size of sample} - 5.113\text{Å}}{5.469\text{Å} - 5.113\text{Å}} \\
 &= \frac{\text{cell size of sample} - 5.113\text{Å}}{0.356\text{Å}}
 \end{aligned}$$

8. Miscellaneous Metallographic Observations

The general integrity of the clad-rib bond was very good. No bondline structures were observable. There was no evidence of intercompartment communication (Figure 20). Regions of fuel-clad interaction, normally noted with this fabrication procedure, were observable throughout all compartments examined (Figures 26C, 27, and 28) and were also found in unirradiated elements (Figures 18 and 31). These oxygen-enriched regions were present at points of maximum stress and in some cases initiated fuel strain cracks which did not progress into the ductile non-interacted Zircaloy cladding.

K. X-Ray Analyses of the Fuel Structures

1. UO₂ Fuel

A section from non-failed UO₂ compartment 1D (Figure 4) was analyzed by X-ray diffractograms to determine the fuel structure. The results of this diffractogram study, shown in Table 8, indicate that there was no shift in lattice parameters for the change in UO₂ cell size in the fluorite-type cubic structure. This is consistent with data from Reference 1.

2. UO₂ + 6 w/o ZrO₂ Fuel

A section from UO₂ + 6 w/o ZrO₂ compartment 2D (Figure 4) was analyzed by using X-ray diffractograms to determine the fuel structure. The results of this diffractogram study are given in Table 8 for comparison with the results from other unirradiated and irradiated UO₂ + 6 w/o ZrO₂ fuels. The cubic cell size of 5.440 ±0.005 Å resolved from sample compartment 2D is comparable to a fuel composition of UO₂ + 4.1 w/o ZrO₂, according to Vegard's Law. The unirradiated control material for Element M22T2 had a cell size of 5.430 ±0.005 Å; this is comparable to a fuel composition of UO₂ + 5.3 w/o ZrO₂. Since there is a cell size tolerance of ±0.005 Å, there is an equivalent ZrO₂ tolerance of ±0.6 w/o. Thus, all numbers are within good agreement. The chemical composition of the unirradiated fuel

TABLE 9. HYDRIDE ANALYSIS FOR STANDARDS AND ELEMENTS M22* AND M21T1

<u>Compartment</u>	<u>Fuel Composition</u>	<u>Sample Weight (g)</u>	<u>Hydrogen Content (ppm)</u>
1A	UO ₂	0.778	53
1D		0.631	60
1E**		0.661	117
1F1		0.275	63
1F2†		0.270	58
2A	UO ₂ + 6 w/o ZrO ₂	0.765	61
2D		0.676	69
2E		0.877	75
2F		0.781	58
M21T1 control	UO ₂	0.568	46
Standard 78	--	0.682	109
Standard 79	--	0.693	109

*The cladding from Element M22 came from nickel-free Zircaloy-2 ingot MS-28349; the hydrogen content was 16 to 19 ppm H₂.

**Failed compartment. This sample was taken from the extreme end of the compartment where a minimum hydrogen content would be expected. Furthermore, the volume of hydride is small when compared with the total volume of the sample.

†The black film on the surface of Element 1F1 was filed off to form sample 1F2.

is also 5.8 w/o ZrO₂. The increase in cell size in the irradiated samples is consistent with the precipitation of excess ZrO₂ noted in Figures 32 and 33. The minimum solubility of ZrO₂ in UO₂ is set by Voronov (Reference 5) at about 5.8 w/o ZrO₂. The precipitation noted in this sample indicates that the solubility limit is probably less than 3.9 w/o. The larger lattice parameters of Element M22T2 should be compared with the lack of change in lattice parameters of Element BHT after irradiation (Table 8).

Element BHT contained a 0.036-inch thickness of fuel comparable to the 0.100-inch thickness in Element M22T2. Hence, Element BHT would probably not have attained temperatures sufficiently high to cause precipitation of excess ZrO₂ as noted in Figure 16A. In addition, the higher fissioning rate of sample BHT may have prevented precipitation as a result of fission-induced phase reversion phenomena.

L. Cladding Hydride Analysis

Eight pieces of Zircaloy cladding from eight fuel compartments of Element M22 were analyzed for hydride content (Figure 4); the results are shown in Table 9. Although these results indicated that the cladding of the failed UO₂ compartment 1E contained an excess of hydrogen compared with that in adjacent compartments, the level of hydrogen did not reflect the high concentrations noted metallographically. It is probable that the cladding section from compartment 1E was cut to contain little of the hydrided region noted in Figures 29 and 30. Hydride contents in unfailed sections of the element show at most a 30-ppm pickup of hydrogen over that of the unirradiated control element. For the weight gain of

81 mg/dm² shown in Figure 10, this yields a percentage pickup of corrosion-produced hydrogen of ~10%.

$$\begin{aligned}\% \text{ hydrogen pickup} &= \frac{\text{observed hydrogen pickup}}{\text{complete hydrogen pickup}} \times 100\% \\ &= \frac{\text{maximum observed hydrogen concentration} \times \text{weight of clad} \times 100\%}{\text{weight gain due to O}_2 \times \text{weight gain of equivalent H}_2},\end{aligned}$$

where

$$\begin{aligned}\frac{\text{weight of clad}}{\text{unit area}} &= \text{density of Zircaloy-2} \times \frac{\text{volume of Zircaloy-2}}{\text{unit area of Zircaloy-2}} \\ &= \text{density of Zircaloy-2} \times \text{clad thickness}.\end{aligned}$$

Therefore,

$$\begin{aligned}\% \text{ hydrogen pickup} &= \frac{30 \times 10^{-6} \frac{\text{gH}_2}{\text{g Zircaloy-2}} \times \frac{6.55 \text{g Zircaloy-2}}{\text{cm}^3} \times 0.02 \text{ in.} \times 2.54 \frac{\text{cm}}{\text{in.}}}{8.1 \frac{\text{gO}_2}{\text{cm}^2} \times \frac{\text{gH}_2}{8\text{gO}_2}} \times 100\% \\ &\cong 10\%\end{aligned}$$

This result is not inconsistent with the hydrogen pickup behavior of nickel-free Zircaloy-2.

M. Emission Spectrographic Analysis of the Cladding

A section of cladding from Element M22 was analyzed for major alloying constituents and for nickel (see Figure 4 for location of sample). The results of the emission spectrographic analysis given in Table 10 show that the nickel content is somewhat higher than that analyzed from nickel-free Zircaloy-2 ingot MS-28349, although it is not large enough to act as a hydrogen sink and cause the catastrophic hydriding present in compartment 1E. The actual nickel content is probably much less than the analyzed values of <100 and <300 ppm; the standard with which these values are compared cannot determine quantities less than 100 ppm of nickel. The pre- and post-irradiation analyses of the chromium content are in very good agreement. However, the tin content in the irradiated material appears to be 50% greater than that from the ingot analysis. This discrepancy cannot be explained at the present time. The iron content, which is the basis for the difference between Zircaloy-2 and Zircaloy-4 (0.12 to 0.18 w/o Fe for nickel-free Zircaloy-2 and 0.18 to 0.24 w/o Fe for Zircaloy-4), in all cases exceeds 0.35 w/o Fe. This iron content is out of the specified range for any known Zircaloy material, but the excessive iron content can be partially explained by the large crud deposits on the surface of Element M22. Although the surface of the emission spectrographic sample was cleaned for one minute with a solution of 50% HNO₃ + 50% H₂O and 5 drops of HF, the surface was probably not completely cleansed of crud deposits. A further analysis check of the major cladding elements will be made at a later time after two mils has been removed from the surface of the spectrographic sample by milling.

III. DISCUSSION OF RESULTS

A. Failure of Element M22

The results of this investigation have shown that Element M22 failed prematurely by a mechanism of accelerated solid volume swelling in the UO₂ fuel. The cause of the accelerated solid volume swelling

TABLE 10. COMPARATIVE EMISSION SPECTROGRAPHIC ANALYSIS OF THE CLADDING OF ELEMENT M22

<u>Sample</u>	<u>Analysis No.</u>	<u>Location</u>	<u>Sn (w/o)</u>	<u>Fe (w/o)</u>	<u>Cr (w/o)</u>	<u>Ni* (ppm)</u>
Ingot MS-28349	--	Top	1.46	0.132	0.115	20
		Middle	1.43	0.128	0.110	20
		Bottom	1.41	0.130	0.1075	20
M21 Control	1	Edge 1	--	0.17	--	<100
	2	Edge 1	--	0.16	--	<100
	3	Side 1	--	0.13	--	<100
	3	Side 2	--	0.13	--	<100
M22**	1	A-1	--	0.39	--	--
	1	A-2	--	0.45	--	--
	1	A-3	--	0.38	--	--
	1	B-4	--	0.47	--	--
	1	B-5	--	0.41	--	--
	1	B-6	--	0.48	--	--
	2	B-1	2.10	0.41	0.12	<300
	2	B-2	2.10	0.50	0.11	<300
	2	B-3	1.90	0.48	0.09	<300
	2	A-4	1.95	0.45	0.11	<300
	2	A-5	2.55	0.35	0.09	<300
	2	A-6	2.50	0.42	0.10	<300
	3	A-1	2.15	0.54	0.13	--
	3	A-2	2.15	0.42	0.11	--

*The nickel content was estimated by comparing standards of 100 ppm and 300 ppm emission spectrographic lines with those from Elements M21 and M22, respectively. The lines from Elements M21 and M22 were too faint to compare with standards representing lower contents. Therefore, the analyzed nickel concentrations are not necessarily the true concentrations.

**Side A: Side opposite that with clad crack and opposite identification (ID) mark.

Side B: Side with clad crack and ID mark.

This sample contained parts of compartments 1F and 2F.

at the premature burnup level was the release of gross amounts of fission gases from small regions of highly fission depleted UO_2 fuel, cracking of the UO_2 matrix, and the subsequent swelling of these regions.

The thermal irradiation history of Element M22 classified this element as a premature failure. Although the heat flux of Element M22 was dropped to extend its life past 17×10^{20} fissions/cc, UO_2 tier M22T1 failed prematurely at 14.7×10^{20} fissions/cc as postulated by the temperature failure curve of Reference 1. The procedure of dropping the heat flux of UO_2 fuel elements and, hence, their fuel centerline temperature has been successful in extending life. Figure 34 illustrates the effects of

Figure 34. Effects of Temperature History on Fuel Element Life for 96% TD UO_2 Fuel.

temperature history on fuel element life. In this figure the actual temperature histories experienced throughout the lifetime of several elements and the time-averaged temperature are plotted as a function of burnup. Elements 2H2 and 9H1, containing 96% TD, 0.040-inch-thick fuel, failed at 31.6×10^{20} and 27.2×10^{20} fissions/cc, respectively, after their heat fluxes had been dropped by 40%. Their time-averaged fuel centerline temperatures were 1860°F (2H2) and 1710°F (9H1). Failure of these elements was anticipated at 19×10^{20} fissions/cc, the failure point for the related Elements 1H2, 2H1, 3H2, 5H1, M31, and M52T1 (Reference 1), if their original heat flux levels had not been lowered. Two other elements, with lowered heat flux and containing 96% TD 0.100-inch-thick UO_2 fuel (7H2) and 0.160-inch-thick UO_2 fuel (8H1), are currently being irradiated as part of the X-1-q test and are past the critical burnup that identical elements 7H1 and 8H2 achieved at their time of failure. Element 7H2 has a current burnup of 11.7×10^{20} fissions/cc and a heat flux of 280,000 Btu/hr-ft² equivalent to a fuel centerline temperature of 1600°F; its sister Element 7H1 failed at 11×10^{20} fissions/cc at a heat flux of 650,000 Btu/hr-ft² (time-averaged temperature was 2900°F). Element 8H1 has a current burnup of 8.0×10^{20} fissions/cc and a heat flux of 580,000 Btu/hr-ft² (3640°F); its sister Element 8H2 failed at 6.2×10^{20} fissions/cc at a heat flux of 670,000 Btu/hr-ft² (3650°F). Another element in the X-3-n test containing 12 compartments of bulk UO_2 fuel, M11 (M11T1 with 96% TD UO_2 and M11T2 with 90% TD UO_2), was recently repositioned in the X-3 loop to lower its heat flux from 380,000 to 240,000 Btu/hr-ft². Element M11T1 is currently at a burnup of 10.7×10^{20} fissions/cc.

Thus, although the heat flux of Element M22 was at the raised level for 42 EFPD prior to its failure, the fuel centerline temperature for Element M22 (M22T1) was not within the lower temperature failure range of pressure-bonded UO_2 fuel elements at any time during this period.

Thus, if both the end-of-life and time-averaged temperatures for Element M22T1 are considered (Figure 34), Element M22 failed prematurely. Furthermore, failed Elements 2H2 and 9H1 and non-failed Elements 7H2 and 8H1 substantiated the theory that the life of an element can be extended by lowering its operating thermal conditions.

The mechanism of failure has been attributed to accelerated solid volume swelling of the UO_2 fuel with verification by the macro-metallographic examination of the fuel-clad interfaces of both the UO_2 and $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuels (Figures 20, 21, 25, and 32) and fission gas release from the $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuels (Figure 12).

Macro-metallographic examination of the fuel-clad interface of three of the most swollen UO_2 compartments including the failed compartment 1E indicated that very little, if any, gas gap was present. The fuel-clad separation accounted for only 6.4% of the total swelling in the non-failed compartments and 10.7% in the failed compartment. Examination of the fuel-clad interface of three $\text{UO}_2 + 6 \text{ w/o ZrO}_2$ fuel compartments indicated that less than 6% of the total compartment swelling was due to the fuel-clad separation. These macro-metallographic observations verify the fact that the fuel was still in an advanced form of the solid volume swelling state.

The fraction of theoretical fission gas release from the six $\text{UO}_2\text{-ZrO}_2$ fuel compartments ranged from 0.0199 to 0.0298. These release figures are in very close agreement with the releases from other $\text{UO}_2\text{-ZrO}_2$ fuels (Reference 2), and verify the fact that no accelerated gas release had commenced within the six M22T2 compartments. On the other hand, comparison of Figures 11 and 12 shows that the UO_2 compartments had an average release substantially greater than that of UO_2 fuels at similar fission depletion levels.

The cause of the large solid volume swelling has, therefore, been attributed to the lack of complete homogenization of the mixtures of $\text{U}^{\text{E}}\text{O}_2$ and $\text{U}^{\text{N}}\text{O}_2$ fuels. This has been verified by the observed swelling of the UO_2 fuel compartments, metallography of the UO_2 fuels, and excessive fission gas release from the UO_2 fuels.

The swelling behavior for the UO_2 fuel material in Element M22 prior to its failure was consistent with that expected for these fuels; this is shown in Figures 7 and 8 by the dotted lines originally from Reference 1. From the comparison of the swelling rates of all six UO_2 fuel compartments and the expected rate of swelling for UO_2 fuel elements, it is evident that the fuels follow the initial swelling rate of about $0.32\% \Delta t$ per 10^{20} fissions/cc ($0.16\% \Delta V$ per 10^{20} fissions/cc). However, at burnup exposures near 11×10^{20} fissions/cc, the departure from the initial swelling rate becomes obvious. This type of behavior has been attributed in the past to the closing of internal pores within the fuel or to the possible formation of a fission gas gap. If the rate of $0.32\% \Delta t / 10^{20}$ fissions/cc is valid for Element M22, then at 14.7×10^{20} fissions/cc the solid volume swelling should have been 14.7×10^{20} fissions/cc $\times 0.32\% \Delta t / 10^{20}$ fissions/cc or $4.7\% \Delta t$ (4.7 mils Δt). The average swelling for the five non-failed UO_2 compartments was $9.5\% \Delta t$, or twice that postulated by the swelling rate (Reference 1). The higher swelling rate experimentally observed is believed related to the microcracking (Figure 22) associated with the highly fission-depleted and grossly swelled $\text{U}^{\text{E}}\text{O}_2$ inhomogeneous regions.

Prior to sintering UO_2 fuels, $\text{U}^{\text{N}}\text{O}_2$ and $\text{U}^{\text{E}}\text{O}_2$ are mixed mechanically to produce the desired U^{235} enrichment. In the case of the fuel used in Element M22, the sintering operation apparently did not completely homogenize the mixture; therefore, isolated regions of $\text{U}^{\text{E}}\text{O}_2$ existed throughout the fuel matrix. This phenomenon was not detected in the unirradiated control samples and was only noted when clumps of fission gas bubbles or "stained" fuel appearances were noticed for irradiated materials. Isolated clumps of $\text{U}^{\text{E}}\text{O}_2$ particles were found in other UO_2 fuels, such as in Elements 2H1 and 7H1 in the X-1-p test (Reference 1), C116 and F13 in the CR-V-m test (Reference 1), and BHU ($\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$, UO_2 enriched to 19.3% U^{235} O_2) in the J-16-1G test (Reference 2). However, the clumps occurred in isolated areas and did not affect the over-all performance of the fuels. The clumps and "stained" fuel materials found within Element M22T1 amounted to 5 to 10% of the total fuel volume; these highly depleted regions, apparently furnished the large release of fission gas mentioned in Section II-H.

If it is assumed that the particles of $\text{U}^{\text{E}}\text{O}_2$ (which after irradiation were observed to be about 45μ in diameter) expanded 60% in volume during irradiation, their original unirradiated diameter was about 28μ . From the observations of microcracks in the matrix of the fuel of Element M22T1 the inpile plasticity of the matrix was insufficient to cause the strains produced at the existing fission depletions.

Not only is the total gas released from each of the five non-failed UO_2 compartments about twice the normal amount, but the variation in fission gas released for each of the five non-failed UO_2 compartments is somewhat higher than that experienced for other UO_2 bearing specimens (Figure 11). These higher release values can be explained by differences in the distribution of highly depleted $\text{U}^{\text{E}}\text{O}_2$ particles in the UO_2 matrix from compartment to compartment. If it is assumed that only one-third of the $\text{U}^{\text{E}}\text{O}_2$ was inhomogeneously distributed and that 5% of the total fuel volume was composed of these highly depleted regions, then 5% of the fuel volume in non-failed UO_2 compartment 1D was depleted to $>90 \times 10^{20}$ fissions/cc and 95% of the fuel volume was depleted to about 10×10^{20} fissions/cc. The percentage of fission gas released from the fuel wafer surface for a fuel exposed to 10×10^{20} fissions/cc is ~1.2% (Figure 11). Since the fission gas release for compartment 1D was 9.2%, the fission gas release from the highly depleted regions was in excess of 25%. This figure of 25% is applicable to fuels exposed to 40×10^{20} fissions/cc and is, therefore, a very conservative estimate for the release from the highly depleted zones.

B. Deterioration of the Cladding by Irradiation and/or Corrosion or Hydriding

Element M22 was irradiated for 478 EFPD, the longest exposure time for any fuel element clad with nickel-free Zircaloy-2 material. The total change in the thickness of failed compartment 1E was 17.7 mils. The five non-failed compartments showed an average total change in thickness of 9.5 mils. Various other UO_2 fuel elements failed in the range from 141 to 262 EFPD, with total fuel compartment thickness increases at the time of failure of 11 to 48 mils. Data for these elements are listed in Table 11 and illustrated in Figure 35. In Figure 35 there is a general pattern of a decrease in cladding ductility as measured by cladding deflection to failure for increasing irradiation time. This loss of ductility is also noted for unirradiated nickel-free Zircaloy-2 elements corrosion tested in out-of-pile autoclaves in 680°F water and 750°F steam (Table 12).

The hydrogen content (24 to 90 ppm) of the unirradiated fuel elements which were corrosion tested in the 680°F water and 750°F steam (Table 12) apparently did not affect the burst strength or ductility of the cladding. Thus, the hydride content (53 to 75 ppm H_2 , Table 9) of the cladding in compartments

TABLE 11. FAILED FUEL COMPARTMENT DEFLECTIONS FOR CERAMIC FUEL ELEMENTS

Test	Element No. (1)	Clad Material	Bond (5)	Fuel Thickness (mils)	EFPD	Failed Compartments	Burnup (fissions/cc $\times 10^{-20}$)	Δt (mils)	% Δt
X-1-p	3L1 ⁽⁴⁾	Nickel-free	IPB	40	260	1C	31	13	32.5
	1H2			40	145	1A	18	37	95.0
	2H1			40	141	2C	17	13	32.5
	2H2			40	197	1C	21	13	32.5
	2H2	Zircaloy-2		40	197	2B	19	23	57.5
	2H2			40	197	2C	21	11	27.5
	3H2			40	148	1C	18	27	67.5
	5H1 ⁽²⁾			40	150	1C	19	31	77.5
	6H2 ⁽³⁾			40	262	1C	31	16	40.0
	7H1			100	231	1A	11	17	17.0
	7H1			100	231	1C	11	48	48.0
	7H1			100	231	2B	10	16	16.0
	7H1			100	231	2C	11	45	45.0
X-3-m	M22	Nickel-free	IPB	100	478	1E	15	18	18.0
	M31			40	162	1D	18	24	60.0
	M52	Zircaloy-2		40	162	1B	20	33	82.5
X-3-1	X1B32	Zircaloy-2	CDB	100	476	1B-1E	14-18	51	51.0
	X1B32			100	476	2B-2E	14-18	24	24.0
	X3N10			40	191	1A	22	26	65.0
	X3N10			40	191	1B-1E	20	34	85.0
	X3N10			40	191	2B-2E	20	34	85.0
	X5N11 ⁽²⁾			40	191	Single comp.	20-22	22	55.0
	X6N12 ⁽²⁾			100	501	Single comp.	15	18	18.0
	X7N11	Zircaloy-4	IPB	100	440	1F-2F	17	76	76.0

TABLE 11 (Cont)

Test	Element No. ⁽¹⁾	Clad Material	Bond ⁽⁵⁾	Fuel Thickness (mils)	EFPD	Failed Compartments	Burnup (fissions/cc $\times 10^{-20}$)	Δt (mils)	% Δt
CR-V-m	I21 ⁽²⁾	Zircaloy-2	CDB	40	401	1	30	18	45.0
	I21 ⁽²⁾			40	401	4	30	20	50.0
	I21 ⁽²⁾			40	401	5	29	16	40.0
	I21 ⁽²⁾			40	401	8	29	14	35.0
	J23			40	406	24	32	16	40.0
VH3-23	W5	Nickel-free Zircaloy-2	IPB	36	107	1D	21	47	130.0
VH3-28	H11	Nickel-free Zircaloy-2	Pack pressure bond	36	163	1A-1C	25	16	44.5
J-16-1H	BFP ⁽⁶⁾	Nickel-free Zircaloy-2	IPB	36	197	F ⁽⁷⁾	23.6	14	35.0
	BFR ⁽⁶⁾	β		36	197	F ⁽⁷⁾	23.6	12	30.0

NOTE: (1) All fuel compartments are 0.250 inch wide and contain 100% UO₂ except where noted.

(2) 0.500-inch wide fuel.

(3) 0.125-inch wide fuel.

(4) Low density fuel (88% TD).

(5) IPB = Isostatic pressure bond.

CDB = Copper diffusion bond (an effective beta treatment).

β = Beta treated.

(6) ZrO₂ + 34 w/o UO₂.

(7) Most swollen non-failed compartment.

Figure 35. Cladding Deflections of Failed Ceramic Fuel Compartments as a Function of Exposure Time.

other than in the failed compartment of Element M22, which may have been present prior to the time of failure and was typical of hydrogen concentrations in Element M22, is not believed to have decreased the cladding ductility appreciably. The catastrophic hydriding observed during destructive evaluation of Element M22 was probably the result and not the cause of the failure. This is consistent with the mechanism proposed in Section III-E.

Table 11 and Figure 35 compare the cladding deflections of failed UO_2 fuel compartments clad with isostatic pressure bonded (IPB) nickel-free Zircaloy-2 and copper diffusion bonded (CDB) Zircaloy-2. The CDB process can be classed as an effective beta treatment since the bonding procedure occurs at 1850-1900°F (the beta treatment is at 1850°F). From the comparison, it is evident that beta-treated clad material can swell twice as much for equal exposure times as non-beta-treated material before the cladding ruptures. The two beta-treated IPB elements shown in Table 11 and Figure 35 are listed for comparison only, since they have not as yet failed. Thus, while the cladding deflection (ductility) of Element M22 was normal for an isostatic pressure-bonded plate, even higher deflections would be anticipated before failure in beta-treated plates.

C. Catastrophic Hydriding

Examination of the failed UO_2 compartment 1E revealed that catastrophic hydriding occurred on the internal clad surface adjacent to the fuel. This phenomenon has been subsequently observed on other failed nickel-free Zircaloy-2 clad fuel elements which were irradiated in the X-1-p and J-16-1J

TABLE 12. EFFECT OF OUT-OF-PILE CORROSION ON SWELLING, HYDROGEN PICKUP, AND PRESSURE BURST PROPERTIES OF INTENTIONALLY DEFECTED ZIRCALOY CLADDING IN PRESSURE BONDED OXIDE PLATES

Corrosion Test (1)	No. of Bonded ⁽²⁾ Plates Tested	No. of Compartments		Exposure (days)	70° F Burst Tested Compartments			H ₂ Content of Defected Clad ⁽⁴⁾ (ppm)
		Per Plate	Defected ⁽³⁾		No.	Rupture Pressure (psi)	Rupture Deflection (mils./side)	
630° F (Water)	14	4	4	129	1	3200	6	46
	8	8	0	129	1	3600	6	34
	14	4	4	101	1	3400	6	36
	8	8	0	101	1	3600	8	33
	14	4	4	73	1	3300	5	49
	8	8	0	73	1	3100	6	36
	2	3	3	65	4	5050-5550	10-35	60-66
	1	3	0	58	2	3650-4050	17-21	39
	14	4	4	56	1	3200	1.6	56
	8	8	0	56	1	3400	6	37
	14	4	4	28	1	4000	8	34
	8	8	0	28	1	4800	14	25
750° F (Steam)	3	8	8	124	1	4000	3.6	90
	3	8	0	124	1	3800	6	65
	3	8	8	96	1	4200	6	71
	3	8	0	96	1	4800	7	47
	3	8	8	56	1	4500	4	58
	3	8	0	56	1	4800	9	39
	3	8	8	28	1	5000	6	34
	3	8	0	28	1	4800	9	24

(1) At 2700 psi.
At 1500 psi.

(2) All cladding made from nickel-free Zircaloy-2. Cladding thickness is 20 mils.
All compartments are 0.250 inch wide x 1.500 inch long.
All fuel is coated with carbon.

(3) Defected compartments have a 0.040-inch diameter defect exposing internal surfaces to coolant.

(4) H₂ content of clad before corrosion testing was 16 to 24 ppm.

TABLE 13. INPILE HISTORY OF FUEL ELEMENTS EXHIBITING INTERNAL HYDRIDING AFTER TIME OF INITIAL FAILURE (ACTIVITY BURST)*

Element	Test	Total EFPD	Time of		Time of Removal		Total Inpile Time after Failure (hours)**
			First Activity Date	Burst Hours	Date	Hours	
2H1	X-1-p	141	11-29-60	1558	11-29-60	1656	1.0
7H1	X-1-p	231	5-9-61	2230	5-29-61	0202	457.7
2H2	X-1-p	197	9-7-61	1700	9-13-61	0223	121.0
M22	X-3-m	478	1-18-62	0430	1-22-62	0201	93.5
	X-3-n						
BHN	J-16-1J	239	8-22-61	1000	10-1-61	2200	911.2

*All fuels were clad with nickel-free Zircaloy-2 material from ingot MS-28349.

**This time has been corrected for trips, shutdowns, and down time in the NRX and NRU reactors.

experiments. These elements, 2H1, 2H2, 7H1, and BHN, all contained 96% TD bulk UO_2 and $\text{UO}_2 + 6 \text{ w/o ZrO}_2$, and were pressure bonded at Battelle in other autoclave bonding runs than that in which Element M22 was bonded. A brief inpile history of these elements after failure and prior to removal is given in Table 13. The inpile time for these elements was from 1 to 911.2 hours. Since Element M22 and the other four elements were bonded at different times and irradiated in different loops, it appears conclusive that the internal hydride is peculiar to the conditions which prevailed in the defective compartments and is not attributable to variables in fabrication procedures or in environmental loop operating conditions.

The hot surface hydriding phenomenon observed in compartment 1E of Element M22 has been analyzed and a reasonable explanation has been hypothesized. Catastrophic hydriding is believed to have occurred for a short time at an extreme pickup rate; during and after this process some hydrogen moved toward the cold surface by the normal thermal diffusion process. To sustain a hydride layer at the hot side during the catastrophic hydriding, a hydrogen flux of $3.3 \text{ mg/cm}^2/\text{day}$ would be required initially, tapering off after saturation of surface film to $1.3 \text{ mg/cm}^2/\text{day}$. To account for the depth of penetration observed, this catastrophic process need only have been sustained for about 21 hours. A mechanism based on the radiolytic decomposition of water trapped in the compartment can account for the necessary hydrogen flux. This preliminary mechanism of hydriding is being extended and developed; it will be reported elsewhere.

D. Corrosion Behavior of Element M22

Examination of the corrosion behavior of the X-3-m and X-3-n specimens listed in Table 14 shows that these specimens, with the exception of Element M52, had average oxide film thicknesses in excess of those calculated from the time and temperature of irradiation. All specimens except Element M52 were pressure bonded at Battelle in autoclave bonding run 1. Element M52 was bonded along with several elements from the X-1-p test (Table 15) in bonding run 2. Table 15 shows that the specimens of the X-1-p test (all samples were fabricated from ingot MS-28349) did not show evidence of accelerated corrosion.

TABLE 14. X-3-M AND X-3-N TEST ELEMENT SURFACE CORROSION AND HYDRIDE BEHAVIOR

Element No.*	Fuel Composition	BMI Bonding Run No.	Avg Inpile Surface Temp (°F)	EFPD	Corrosion Film (μ)			Localized Heavy Hydride at Surface	Discharge Date	Cause for Discharge
					Measured Max.	Avg	Calc Avg			
M11	UO ₂	1	--	--	--	--	--	--	Inpile, X-3-n test	--
M22	UO ₂ ; UO ₂ + 6 w/o ZrO ₂	1	560	479	24	7	1.6	Yes-at failure	1-22-62	UO ₂ Comp. 1E swelled and leaked.
M31	UO ₂	1	590	162	7	3	1.4	Yes-at failure	11-21-60	Comp. 1D swelled and leaked.
M42	UO ₂	1	580	338	13	8	1.6	Yes	7-24-61	Gross swelling in all twelve compartments
M52	UO ₂ ; UO ₂ + 6 w/o ZrO ₂	2	591	162	3	2	1.4	No	11-21-60	UO ₂ Comp. 1A grossly swelled; UO ₂ Comp. 1B swelled and leaked.
M61	UO ₂ + 2.1 and 4.9 w/o Y ₂ O ₃	1	--	358	Not yet examined				3-19-62	Gross swelling in all four compartments
M72	UO ₂	1	590	133	>200	2	1.3	Yes-at failure	4-17-61	"Burnout" in Comp. 1C
15D	UO ₂ + 65 w/o ZrO ₂	15	--	--	--	--	--	--	Inpile, X-3-n test	--
B72	UO ₂ + 53 w/o ZrO ₂	5	--	--	--	--	--	--	Inpile, X-3-n test	--
2L1- β	UO ₂	2	--	--	--	--	--	--	Inpile, X-3-n test	--

*All elements were clad with material from nickel-free Zircaloy-2 ingot MS-28349. They were all isostatically pressure bonded at 1550°F for 4 hours. Elements indicated with (- β) were beta treated.

**All non-beta treated elements were given a pre-irradiation corrosion test in 680°F H₂O for 6 days. All beta-treated elements were given a pre-irradiation corrosion test in 680°F H₂O for 3 days.

TABLE 15. X-1-P AND X-1-Q TEST UO₂ ELEMENT SURFACE CORROSION AND HYDRIDE BEHAVIOR

Element ⁽¹⁾ No.	BMI Bonding Run No.	Avg Inpile Surface Temp (°F)	EFPD	Corrosion Film (μ) ⁽²⁾			Localized Heavy Hydride at Surface	Discharge Date	Cause for Discharge
				Measured Max.	Avg	Calc Avg			
1L1	2	568	259	3	1	1.4	No	7-7-61	Gross swelling
1L2	2	577	261	Not yet examined		1.5	--	1-15-62	Gross swelling
2L1-β	2	--	--	--	--	--	--	Inpile, X-3-n test	--
2L2	2	587	141	Not yet examined		1.5	--	11-29-60	No failure
3L1	2	573	259	--	<1	1.5	Yes-at failure	7-7-61	Comp. 1C swelled and leaked
3L2-β	2	--	--	--	--	--	--	Inpile, X-1-q test	--
4L1	2	587	139	--	<1	1.5	No	11-21-60	No failure
4L3-β	11	--	--	--	--	--	--	Inpile, X-1-q test	--
1H2	2	593	145	--	<1	1.6	No	12-19-60	Comp. 2A swelled and leaked
2H1	2	593	141	--	1	1.6	Yes-at failure	11-29-60	Comp. 2C swelled and leaked
2H2	2	579	197	--	<1	1.4	Yes-at failure	9-13-61	Comps. 1C, 2B, and 2C swelled and leaked.
3H2	2	592	148	Not yet examined		1.5	--	1-22-61	Comp. 1C swelled and leaked
4H2	3	594	139	Not yet examined		1.5	--	11-21-60	No failure
5H1	3	592	150	Not yet examined		1.6	--	1-22-61	Comp. C swelled and leaked
6H2	4	580	261	6	1.5	1.5	Yes-at failure	7-7-61	Comp. 1C swelled and leaked
7H1	3	593	231	--	2	1.7	Yes-at failure	5-29-61	Comps. 1A, 1C, 2B and 2C swelled and leaked.
7H2	3	--	--	--	--	--	--	Inpile, X-1-q test	--
8H1	4	--	--	--	--	--	--	Inpile, X-1-q test	--
8H2	4	596	198	--	<1	1.6	Yes-at failure	4-17-61	Gross swelling of Comps. 1C and 2C
9H1	3	568	288	Not yet examined		1.4	--	1-15-62	Comps. 1C, 1B, and 2C swelled and leaked.

(1) All elements were clad with material from nickel-free Zircaloy-2 ingot MS-28349. They were all isostatically pressure bonded at 1550°F for 4 hours. Elements indicated with (-β) were beta quenched.

(2) All non-beta-treated elements were given a pre-irradiation corrosion test in 680°F H₂O for 6 days. All beta-treated elements were given a pre-irradiation corrosion test in 680°F H₂O for 3 days.

TABLE 16. J-16 CERAMIC SEED ELEMENTS SHOWING SEVERE WHITE CORROSION

Element*	Fabrication No.	BMI Bonding Run No.	Average Inpile Surface Temp (° F)	EFPD	Corrosion Film (μ)**			Localized Heavy Hydride at Surface†
					Measured Max.	Avg	Calculated Avg	
BFY	17A	17	623	147	--	--	--	No
BHA	17C	17	627	147	24	3	1.7	No
BFZ	17B	17	625	147	30	1	1.6	No
BHN	21C	21	--	239	--	--	--	Yes
BHO	22A	22	--	239	--	--	--	No
BHD	19A	19	624	197	--	--	--	No
BHH	19D	19	628	197	--	--	--	No

*All elements were clad with material from nickel-free Zircaloy-2 ingot MS-28349, and were all isostatically pressure bonded at 1550°F for 4 hours. All elements contained six compartments of $\text{ZrO}_2 + 35 \text{ w/o } \text{UO}_2$ ceramic seed fuel.

**All elements were given a pre-irradiation corrosion test in 680° F H_2O for 6 days.

†No J-16 elements have been found with internal hydrides except for Element BHN.

Table 16 lists some pertinent information from the J-16 test. Here, too, the specimens were prepared by pressure bonding at BMI without remedial beta treatment of clad material from ingot MS-28349. The seven specimens showing accelerated corrosion, again, were from four Battelle pressure-bonding runs. The J-16 test contained specimens from about seven other pressure-bonding runs, and none of these specimens showed accelerated corrosion.

Thus, it appears that the accelerated corrosion observed on X-3-m and J-16 specimens is a consequence of pressure bonding nickel-free Zircaloy-2 without remedial beta treatment, and may also be related specifically to factors (unknown at present) existing in the pressure-bonding autoclaves during particular bonding operations. Some support for this conclusion is offered by the out-of-pile corrosion testing of J-16 companion samples at 750° F (Table 17). Table 17 shows that, in general, the corrosion rate of these specimens is high in 750° F steam and normal in 680° F water. In general, the corrosion results are entirely consistent with the behavior of nickel-free Zircaloy-2 subjected to pressure bonding. From this, it may be interred that the specimens of the X-3-m tests did not behave unexpectedly out-of-pile in respect to cladding corrosion. With the exception of Elements M22, 2H1, 2H2, 7H1, and BHN, all specimens of the X-3-m, X-1-p, and J-16 tests showed normal hydriding behavior; i.e., where local hydriding occurred, it was at those cooler locations of the external clad surface of the specimen where hydride concentration would be expected. In particular, in the J-16 test where corrosion on a generally larger scale took place and large quantities of hydrogen were available, no peculiar distribution of hydride was observed other than that in Element BHN.

Therefore, although the abnormal corrosion observed on the subject specimens was undoubtedly related to the prior fabrication history of the fuel elements, the origin and distribution of hydride in Element M22 (and in Elements 2H1, 2H2, 7H1, and BHN) lay elsewhere and was not a consequence of corrosion, since specimens from other tests, notably J-16, would be expected to show the same

TABLE 17. AUTOCLAVED OUT-OF-PILE COMPANIONS TO IRRADIATION PLATES
CLAD WITH NICKEL-FREE ZIRCALOY-2 FROM INGOT MS-28349

Out-of-Pile Specimen	Inpile Companion	Irradiation Test	Fuel	Corrosion* Test Temp	Maximum Exposure Time	Remarks
M32	M31**	X-3-m	UO ₂	680° F (W)	--	Still in first 28-day exposure period.
M33	M31**	X-3-m	UO ₂	750° F (S)	--	Still in first 28-day exposure period.
2OD†	BHJ, BEK	J-16	ZrO ₂ + 35 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film.
4C	BFD	J-16	ZrO ₂ + 25 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film.
8A	BJB, BJC	J-16	ZrO ₂ + 18 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film.
9A	BHX	J-16	ZrO ₂ + 80 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film.
3A	BHV	J-16	ZrO ₂ + 87 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film, one spot near stamped numbers.
6D	BHY, BHZ	J-16	ZrO ₂ + 60 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film.
5A	BJD	J-16	ZrO ₂ + 25 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film.
B61	--	J-16	ZrO ₂ + 34 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film, small spots on both sides of samples.
7C	BFW	J-16	ZrO ₂ + 20 w/o UO ₂	680° F (W)	42 days	Lustrous black corrosion film.
4A	BFT	J-16	ZrO ₂ + 34 w/o UO ₂	750° F (S)	42 days	Welds very bad, tan oxide over all surfaces: ΔW ~50 mg (~100 mg/dm ²).
10A	1ZA	J-16	ZrO ₂ + 34 w/o UO ₂	750° F (S)	42 days	Welds are tan, grey-tan oxide over all surfaces: ΔW ~50 mg (~100 mg/dm ²).
21A	BHM, BHN	J-16	ZrO ₂ + 34 w/o UO ₂	750° F (S)	42 days	Welds are tan, grey-tan oxide over all surfaces: ΔW ~40 mg (~80 mg/dm ²).
8C	BJC	J-16	ZrO ₂ + 18 w/o UO ₂	750° F (S)	42 days	One connector tan, welds are tan, tan surfaces: ΔW ~70 mg (~140 mg/dm ²).
9D	BHW	J-16	ZrO ₂ + 87 w/o UO ₂	750° F (S)	42 days	Tan welds, grey-tan oxide over all surfaces: ΔW ~70 mg (~140 mg/dm ²).
3C	BHT	J-16	ZrO ₂ + 94 w/o UO ₂	750° F (S)	42 days	Tan welds, grey-tan oxide over all surfaces: ΔW ~50 mg (~100 mg/dm ²).
6C	BHY	J-16	ZrO ₂ + 60 w/o UO ₂	750° F (S)	42 days	Tan welds, grey-tan oxide over all surfaces, one spot on non-numbered side: ΔW ~50 mg (~100 mg/dm ²).
7B	BHU	J-16	ZrO ₂ + 94 w/o UO ₂	750° F (S)	42 days	Tan welds, grey-tan oxide over all surfaces, one tan end connector: ΔW ~70 mg (~140 mg/dm ²).

TABLE 17 (Cont)

Out-of-Pile Specimen	Inpile Companion	Irradiation Test	Fuel	Corrosion* Test Temp	Maximum Exposure Time	Remarks
B62	--	J-16	ZrO ₂ + 34 w/o UO ₂	750° F (S)	42 days	Tan welds, grey-tan oxide over all surfaces: $\Delta W \sim 50$ mg (~ 100 mg/dm ²).
17	--	J-16	--	750° F (S)	70 days	Grey-tan "splotchy" oxide over all surfaces: $\Delta W = 112$ mg/dm ² .
18	--	J-16	--	750° F (S)	70 days	Grey-tan "splotchy" oxide over all surfaces: $\Delta W = 108$ mg/dm ² .
19	--	J-16	--	750° F (S)	70 days	Grey-tan "splotchy" oxide over all surfaces: $\Delta W = 109$ mg/dm ² .
20	--	J-16	--	750° F (S)	70 days	Grey-tan "splotchy" oxide over all surfaces: $\Delta W = 112$ mg/dm ² .
22	--	J-16	--	750° F (S)	70 days	Grey-tan "splotchy" oxide over all surfaces: $\Delta W = 114$ mg/dm ² .

*(W) = water at 2700 psi.

(S) = steam at 1500 psi.

**Elements M32 and M33 are controls for all X-1-p and X-3-m elements.

†Beta-treated.

phenomenon. The source of hydrogen and distribution of hydrides in Element M22 must, therefore, represent conditions and factors peculiar to that specimen.

In Figure 10 the rate of weight gain for Element M22 has two definite patterns. One pattern of $0.24 \text{ mg/dm}^2/\text{day}$ is that generated prior to the removal of intentionally defected Element X7N11 from the X-3 loop on April 17, 1961. The other pattern of $0.085 \text{ mg/dm}^2/\text{day}$ is that generated after the removal of defected element X7N11. The second pattern is similar to that found for elements in the X-1 loop (superimposed on Figure 10). Evidently, the initial high corrosion weight gain of Element M22 was due to the presence of uranium from defected Element X7N11 in the X-3 loop.

Corrosion behaviors demonstrated by Element M22 are as follows:

1. The element had an inpile corrosion rate characteristic of Zircaloy fabricated in the $\gamma + \beta$ range of temperatures.
2. The bonding run in which the element was fabricated deteriorated its corrosion resistance more than would be anticipated from the pressure-bonding operation.
3. Large amounts of uranium in the X-3 loop during a part of the life of Element M22 also affected the corrosion behavior.
4. The cracking of the cladding and/or presence of localized regions of coarse grain size further affected local corrosion resistance.

In spite of these anomalies cladding corrosion was apparently not a significant factor in specimen failure.

IV. CONCLUSIONS

The failure of Element M22 is attributed to the abnormally large solid volume swelling of the UO_2 fuel in compartment 1E rather than to the formation of an insulating fission gas gap. The cause of the abnormally large solid volume swelling was the large amount of fission released from and the subsequent swelling of highly depleted pockets (burnups in excess of 90×10^{20} fissions/cc) of $\text{U}^{\text{E}}\text{O}_2$ distributed within the UO_2 matrix. The pockets were attributed to regions of inhomogeneous mixtures of $\text{U}^{\text{E}}\text{O}_2$ in $\text{U}^{\text{N}}\text{O}_2$.

Other major conclusions are as follows:

1. The corrosion behavior of Element M22 was that expected for an X-3 loop element.
2. Catastrophic hydriding of the internal clad surfaces was the result and not the cause of fuel element failures. A mechanism based on the radiolytic decomposition of water trapped within a compartment after failure can account for the necessary hydrogen flux to initiate and maintain the hydride structure in a hot region of cladding.
3. The ductility of the nickel-free Zircaloy-2 cladding decreased as the irradiation exposure time increased. This decrease was probably caused by irradiation damage, with some small contributions by water corrosion and hydrogen pickup.
4. Beta-treated Zircaloy (nickel-free Zircaloy-2 and Zircaloy-4) displayed higher ductility and higher deflection as a function of inpile exposure time than did non-beta-treated oxide plates such as those in Element M22.
5. The $\text{UO}_2 + 6 \text{ w/o } \text{ZrO}_2$ fuels released expected amounts of fission gas and did not undergo any visible grain size changes in regions near the fuel-clad interface. The fuel, however, contained precipitates, presumably of a ZrO_2 -rich phase. The swelling of these fuels was similar to the swelling of fuels previously irradiated (reported in Reference 2).

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APPENDIX A
INTERIM EXAMINATION DATA FOR ELEMENT M22

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TABLE A-1. INTERIM EXAMINATION DATA (AT CHALK RIVER)

Location	Bettis Original	Thickness Measurements (in.)						
		Exam. No. 1 7-26-60	Exam. No. 2 9-19-60	Exam. No. 3 11-21-60	Exam. No. 4 12-19-60	Exam. No. 5 1-24-61	Exam. No. 6 2-24-61	Exam. No. 7 4-19-61
1A1	0.1411	0.1406	0.1410	0.1424	0.1429	0.1420	0.1428	0.1438
1A2	0.1398	0.1404	0.1406	0.1421	0.1425	0.1420	0.1428	0.1437
1A3	0.1412	0.1407	0.1407	0.1426	0.1428	0.1419	0.1430	0.1437
1B1	0.1407	0.1403	0.1406	0.1420	0.1424	0.1418	0.1430	0.1435
1B2	0.1411	0.1407	0.1406	0.1423	0.1423	0.1420	0.1428	0.1436
1B3	0.1405	0.1406	0.1408	0.1422	0.1421	0.1425	0.1426	0.1431
1C1	0.1412	0.1403	0.1407	0.1422	0.1422	0.1426	0.1427	0.1433
1C2	0.1405	0.1401	0.1401	0.1415	0.1415	0.1419	0.1425	0.1429
1C3	0.1406	0.1403	0.1406	0.1423	0.1418	0.1423	0.1428	0.1430
1D1	0.1412	0.1406	0.1409	0.1425	0.1425	0.1428	0.1433	0.1434
1D2	0.1414	0.1409	0.1414	0.1430	0.1428	0.1429	0.1434	0.1434
1D3	0.1411	0.1408	0.1409	0.1425	0.1422	0.1428	0.1428	0.1433
1E1	0.1411	0.1409	0.1415	0.1430	0.1430	0.1430	0.1435	0.1439
1E2	0.1407	0.1405	0.1408	0.1425	0.1424	0.1426	0.1424	0.1430
1E3	0.1411	0.1408	0.1410	0.1429	0.1428	0.1430	0.1434	0.1437
1F1	0.1404	0.1406	0.1406	0.1423	0.1422	0.1427	0.1427	0.1431
1F2	0.1403	0.1404	0.1410	0.1422	0.1420	0.1423	0.1423	0.1428
1F3	0.1413	0.1406	0.1412	0.1422	0.1421	0.1420	0.1429	0.1430
2A1	0.1401	0.1408	0.1415	0.1432	0.1430	0.1433	0.1433	0.1435
2A2	0.1404	0.1409	0.1416	0.1426	0.1428	0.1434	0.1433	0.1436
2A3	0.1406	0.1404	0.1408	0.1420	0.1420	0.1425	0.1427	0.1430
2B1	0.1401	0.1405	0.1410	0.1420	0.1421	0.1426	0.1430	0.1431
2B2	0.1402	0.1407	0.1415	0.1429	0.1425	0.1430	0.1437	0.1438
2B3	0.1408	0.1408	0.1416	0.1430	0.1429	0.1431	0.1431	0.1434
2C1	0.1396	0.1408	0.1410	0.1425	0.1423	0.1427	0.1430	0.1433
2C2	0.1397	0.1408	0.1410	0.1428	0.1425	0.1427	0.1430	0.1431
2C3	0.1401	0.1405	0.1411	0.1425	0.1421	0.1425	0.1429	0.1432
2D1	0.1396	0.1402	0.1408	0.1429	0.1430	0.1428	0.1430	0.1439
2D2	0.1400	0.1405	0.1409	0.1429	0.1432	0.1429	0.1431	0.1444
2D3	0.1406	0.1401	0.1401	0.1422	0.1421	0.1433	0.1427	0.1428
2E1	0.1393	0.1402	0.1413	0.1425	0.1431	0.1425	0.1431	0.1437
2E2	0.1400	0.1407	0.1418	0.1430	0.1432	0.1430	0.1434	0.1432
2E3	0.1401	0.1405	0.1416	0.1430	0.1430	0.1429	0.1431	0.1427
2F1	0.1387	0.1405	0.1413	0.1430	0.1432	0.1431	0.1431	0.1431
2F2	0.1401	0.1407	0.1415	0.1429	0.1431	0.1431	0.1434	0.1431
2F3	0.1398	0.1408	0.1414	0.1423	0.1424	0.1426	0.1428	0.1432

TABLE A-1 (Cont)

Location	Thickness Measurements (in.)						Bettis 2-9-62
	Exam. No. 8 6-26-61	Exam. No. 9 7-24-61	Exam. No. 10 9-18-61	Exam. No. 11 10-23-61	Exam. No. 12 11-21-61	Exam. No. 13 1-22-62	
1A1	0.1458	0.1460	0.1469	0.1483	0.1480	0.1502	0.1498
1A2	0.1452	0.1458	0.1461	0.1465	0.1466	0.1491	0.1487
1A3	0.1460	0.1460	0.1467	0.1480	0.1489	0.1502	0.1490
1B1	0.1450	0.1453	0.1460	0.1472	0.1471	0.1499	0.1488
1B2	0.1451	0.1454	0.1463	0.1471	0.1469	0.1492	0.1487
1B3	0.1450	0.1450	0.1465	0.1468	0.1469	0.1497	0.1489
1C1	0.1450	0.1452	0.1462	0.1469	0.1453	0.1469	0.1486
1C2	0.1440	0.1448	0.1451	0.1456	0.1458	0.1485	0.1478
1C3	0.1442	0.1450	0.1453	0.1460	0.1472	0.1483	0.1476
1D1	0.1450	0.1455	0.1465	0.1471	0.1469	0.1500	0.1530
1D2	0.1451	0.1455	0.1462	0.1470	0.1470	0.1550	0.1510
1D3	0.1450	0.1450	0.1462	0.1467	0.1468	0.1545	0.1514
1E1	0.1450	0.1454	0.1462	0.1475	0.1473	0.1513	0.1575
1E2	0.1450	0.1452	0.1461	0.1470	0.1469	0.1583	0.1584
1E3	0.1452	0.1454	0.1471	0.1475	0.1475	0.1595	0.1580
1F1	0.1442	0.1449	0.1458	0.1465	0.1455	0.1455	0.1501
1F2	0.1443	0.1448	0.1462	0.1468	0.1464	0.1510	0.1511
1F3	0.1447	0.1455	0.1460	0.1465	0.1458	0.1508	0.1505
2A1	0.1455	0.1459	0.1462	0.1465	0.1468	0.1489	0.1484
2A2	0.1451	0.1452	0.1461	0.1465	0.1463	0.1484	0.1478
2A3	0.1442	0.1443	0.1453	0.1456	0.1461	0.1472	0.1460
2B1	0.1445	0.1448	0.1450	0.1453	0.1448	0.1465	0.1460
2B2	0.1449	0.1452	0.1455	0.1460	0.1457	0.1470	0.1470
2B3	0.1449	0.1452	0.1455	0.1460	0.1458	0.1470	0.1468
2C1	0.1441	0.1447	0.1450	0.1452	0.1436	0.1458	0.1457
2C2	0.1445	0.1451	0.1452	0.1453	0.1453	0.1470	0.1465
2C3	0.1442	0.1445	0.1455	0.1457	0.1455	0.1470	0.1464
2D1	0.1442	0.1450	0.1455	0.1455	0.1454	0.1472	0.1462
2D2	0.1448	0.1451	0.1455	0.1458	0.1459	0.1472	0.1467
2D3	0.1441	0.1445	0.1455	0.1455	0.1458	0.1471	0.1461
2E1	0.1445	0.1449	0.1459	0.1463	0.1432	0.1479	0.1470
2E2	0.1449	0.1459	0.1459	0.1462	0.1464	0.1480	0.1474
2E3	0.1444	0.1450	0.1457	0.1460	0.1462	0.1473	0.1466
2F1	0.1445	0.1450	0.1459	0.1469	0.1466	0.1457	0.1474
2F2	0.1450	0.1451	0.1460	0.1465	0.1467	0.1485	0.1479
2F3	0.1447	0.1452	0.1453	0.1468	0.1465	0.1485	0.1483

TABLE A-1 (Cont)

<u>Location</u>	<u>Bettis Original</u>	<u>Exam. No. 1 7-26-60</u>	<u>Exam. No. 2 9-19-60</u>	<u>Exam. No. 3 11-21-60</u>	<u>Exam. No. 4 12-19-60</u>	<u>Exam. No. 5 1-24-61</u>	<u>Exam. No. 6 2-24-61</u>	<u>Exam. No. 7 4-19-61</u>
<u>Width Measurements (in.)</u>								
W1	0.8135	0.8112	0.8128	0.8128	0.8124	0.8137	0.8136	0.8140
W2	0.8130	0.8111	0.8119	0.8133	0.8139	0.8131	0.8142	0.8133
W3	0.8125	0.8100	0.8107	0.8112	0.8139	0.8132	0.8110	0.8130
W4	0.8125	0.8094	0.8108	0.8100	0.8112	0.8112	0.8107	0.8112
W5	0.8126	0.8100	0.8102	0.8103	0.8135	0.8119	0.8110	0.8133
W6	0.8124	0.8101	0.8105	0.8114	0.8119	0.8121	0.8119	0.8126
W7	0.8130	0.8112	0.8119	0.8128	0.8115	0.8126	0.8124	0.8137
W8	0.8145	0.8104	0.8104	0.8122	0.8110	0.8118	0.8118	0.8121

Length Measurements (in.)

Length (max.)	11.159	--	--	--	--	--	11.1640	11.163
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Dry Weight Measurements (g)

Weight	160.8790	160.8896	160.9179	160.9334	160.9426	160.9497	160.9567	160.9600
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Density Measurements (g/cc)

Density	7.92	--	--	--	--	--	--	--
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Bow Measurements (+ in.)

Bow	0.018	--	--	--	--	--	--	0.045
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<u>Location</u>	<u>Exam. No. 8 6-27-61</u>	<u>Exam. No. 9 7-24-61</u>	<u>Exam. No. 10 9-18-61</u>	<u>Exam. No. 11 10-23-61</u>	<u>Exam. No. 12 11-21-61</u>	<u>Exam. No. 13 1-22-62</u>	<u>Bettis 2-9-62</u>
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Width Measurements (in.)

W1	0.8131	0.8131	0.8131	0.8132	0.8131	0.8140	0.8139
W2	0.8139	0.8141	0.8131	0.8138	0.8138	0.8135	0.8143
W3	0.8113	0.8129	0.8132	0.8133	0.8116	0.8120	0.8132
W4	0.8106	0.8126	0.8128	0.8128	0.8102	0.8112	0.8132
W5	0.8111	0.8123	0.8128	0.8132	0.8112	0.8113	0.8131
W6	0.8120	0.8122	0.8129	0.8128	0.8115	0.8123	0.8125
W7	0.8121	0.8131	0.8132	0.8135	0.8132	0.8133	0.8140
W8	0.8119	0.8117	0.8127	0.8130	0.8138	0.8140	0.8153

Length Measurements (in.)

Length (max.)	11.162	11.162	11.162	11.164	11.162	11.167	11.1672
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TABLE A-1 (Cont)

<u>Location</u>	<u>Exam. No. 8 6-27-61</u>	<u>Exam. No. 9 7-24-61</u>	<u>Exam. No. 10 9-18-61</u>	<u>Exam. No. 11 10-23-61</u>	<u>Exam. No. 12 11-21-61</u>	<u>Exam. No. 13 1-22-62</u>	<u>Bettis 2-9-62</u>
<u>Dry Weight Measurements (g)</u>							
Weight	160.9668	160.9670	160.9743	160.9768	160.9808	160.9847	--
<u>Density Measurements (g/cc)</u>							
Density	--	--	--	--	7.7917	--	--
<u>Bow Measurements (+ in.)</u>							
Bow	--	--	--	--	--	0.044	--

APPENDIX B*

OPERATING HISTORY FOR THE X-3-m AND X-3-n
TESTS DURING IRRADIATION OF ELEMENT M22
(MAY 6, 1960, TO FEBRUARY 13, 1962)

*The information in this appendix was supplied by R. T. Esper.

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A. Introduction

This appendix summarizes the operating history for the X-3-m and X-3-n tests conducted in the NRX X-3 loop at Chalk River. Specifically, loop operating levels, coolant chemistry conditions, and radiochemical data covering the period from the start of the X-3-m test on May 6, 1960, until shortly after failure of specimen M-22 in the X-3-n test on January 18, 1962, are reviewed and discussed. The gross activity bursts during test operation and the causes of the subsequent high activity levels are also presented in detail.

B. Test History

The X-3-m test started operation in the NRX X-3 loop on May 6, 1960. The specimen assembly for this test included one defected specimen previously irradiated in the X-3-l test. The specimen, identified as Element X7N11, had a 0.040-inch defect hole. During this test six specimens were removed because of failure; at times the failed specimen was replaced with another specimen. The test ended on November 20, 1961. At that time the X-3-m irradiated test specimens were rearranged and new, unirradiated specimens were added. The new test specimen assembly was identified as the X-3-n test which started operation on November 26, 1961. Although this test continues in operation at the present time, the information in this appendix covers the period to February 13, 1962, or to a point approximately one month after the failure of Element M22.

C. Loop Operation

The design operating and coolant chemistry conditions for the X-3-m and X-3-n tests in addition to the actual operating levels are summarized in Table B-1. Throughout the entire period of operation for both the X-3-m and X-3-n tests the loop operating conditions were maintained at specified pressure and flowrate levels. The bulk water inlet temperature range for the X-3-m test was 485 to 495°F; for the X-3-n test the range was 460 to 465°F. During the life of both tests no excursion in temperature, pressure, or flowrate levels was observed; nor were any significant loop mechanical difficulties encountered.

D. Coolant Chemistry Conditions

The loop water chemical conditions for the X-3-m and X-3-n tests were carefully controlled and steadily monitored. The range of hydrogen concentration during the X-3-m test was 10 to 35 cc/kg and during the X-3-n test was 11 to 20 cc/kg. Hydrogen additions were made frequently to the system during operation to maintain specified levels. The oxygen concentration in both tests was significantly less than the 0.05-ppm maximum requirement.

The coolant pH was maintained by the use of LiOH-type mixed bed, ion exchange resin during the life of both tests. During the X-3-m test the pH level ranged from 9.5 to 10.88, with an average of approximately 10.2 for most of the run. The pH range during the X-3-n test was 9.5 to 10.1, with an average of 9.9. The test pH levels were well within specified limits.

At the start of the X-3-m test a coolant crud level value of 0.02 ppm was determined. The value determined a week later was less than 0.008 ppm. Since that time and until the end of this report period the crud concentration levels in the X-3 loop have been less than 0.10 ppm. A crud sample was analyzed prior to each monthly loop startup.

E. Radiochemical Conditions

With the start of the X-3-m test on May 6, 1960, leaching of the UO_2 from the 0.040-in. defect hole in Element X7N11 caused an increase in loop activity levels. The activity maintained a rather constant level and loop operation continued in a satisfactory manner. The 33m Cs^{138} values ranged from 2×10^6 to 9×10^6 dpm/ml, and the range of 2.8h Kr^{88} levels was 2×10^6 to 2×10^7 dpm/ml. A slight gradual increase in the specific activity levels was observed during late October and early November of 1960. At approximately 2030 on November 11, 1960, the DN and gamma monitors in the X-3 loop showed a gross increase in activity levels. Radiochemical analyses verified the activity increase. The test was removed for a detailed examination on November 21, 1960. The examination revealed the failure of two previously sound elements, M31 and M52. These elements were removed from test and replaced with Elements M72 and M61. Defected Element X7N11 was continued in test.

The X-3-m test operation continued in a satisfactory manner, with Cs^{138} levels ranging from 2×10^6 to 6×10^6 dpm/ml and Kr^{88} levels ranging from 2×10^6 to 7×10^6 dpm/ml. Suddenly, at 1325 on January 18, 1961, a substantial increase in coolant activity was detected and subsequently verified by radiochemical analyses. The test was removed from the reactor during the January 23, 1961, shutdown. An examination revealed the failure of Element X6N12, which was subsequently removed from test. The test assembly was reassembled and returned for continued irradiation.

Operation of the test continued without incident after reactor startup on January 26, 1961, with specific activity levels in the same range as previously indicated. On February 17, 1961, at 0405, the gamma monitor went off scale abruptly at its maximum range, indicating a specimen failure. Standard activity samples taken shortly thereafter showed a substantial increase over steady-state survey values. The NRX reactor had been at full and steady power and the loop at normal operating conditions. During the following 12 hours the loop activity as measured by the gamma monitor appeared to be decreasing at a slow rate; however, at approximately 1600 the gamma monitor again went off scale. Loop water samples were too active to analyze. Loop radiation surveys showed a substantial increase in activity above normal observed levels. The reactor was shut down on February 17, 1961, and the test was removed and examined. Failure of non-defected Element X1B32 was the source of high activity. This specimen was removed from test.

The experimental assembly was returned to the reactor, and test performance was comparatively normal, with the specific activity levels remaining within the ranges mentioned until approximately 0500 on April 16, 1961. At that time the loop radiation survey increased greatly over normal levels. Activities of standard water samples also increased substantially over normal levels. The NRX reactor had been at full and steady power since 0600 on April 15, 1961. Specific activity analyses indicated that on April 17, 1961, the gross $\beta - \gamma \text{Cs}^{138}$, Kr^{88} , and standard sample activities had increased over normal levels substantially. The NRX reactor was shut down at 2224 on April 17, 1961, and the test was removed for examination, which revealed the failure of non-defected element M72 and the failure of two non-defected compartments in Element X7N11. Both specimens were removed from test and the experiment was returned for further irradiation. During the July 24, 1961, shutdown the X-3-m test was rearranged to include irradiated Elements 15D and B72, which were previously removed from the J-16-1G test. At the same time, Element M42 was removed because of excessive fuel compartment swelling.

Subsequent to the removal of defective specimens M72 and X7N11 and until the end of the X-3-m test on November 20, 1961, the coolant Cs^{138} , Kr^{88} , gross $\beta - \gamma$, and gross I levels decreased considerably from levels previously observed. This period of operation showed Cs^{138} levels ranging from 1.0×10^6 to 1.8×10^6 dpm/ml and Kr^{88} levels within the range of 1.1×10^6 to 1.9×10^6 dpm/ml. The levels continued to decrease after the removal of the X-3-m test and the insertion of the X-3-n test on November 20, 1961.

At 0430 on January 18, 1962, an initial burst of activity was detected in the X-3 loop. The gamma monitor read off scale and the activity level of a standard water sample reached 1R. Radiochemical analysis of the loop water verified the high loop activity and indicated a specimen failure. However, the test remained inpile, and the activity levels decreased gradually as a result of purification system cleanup until January 22, 1962, when the test assembly was removed and examined. The examination revealed a failure in Element M22, which was subsequently removed from test. The specimen train was reassembled and returned to the reactor for continued irradiation.

In the various specimen failures discussed, coolant activity increased suddenly and without prior indication. In Table B-2, activity levels for each failure are presented for the periods before and after the activity burst. As evidenced from this data, failure of a specimen is characterized by a sudden surge of activity levels.

TABLE B-1. LOOP OPERATING AND CHEMISTRY CONDITIONS

Date	X-3-m Test		X-3-n Test	
	5-6-60 to 11-20-61		11-20-61 to 2-18-62	
Data	Design	Actual	Design	Actual
Flowrate (USGPM)	12	12	10	10
Pressure (psi)	2000	2000	2000	2000
T _{Inlet} (°F)	490	485-495	460	460-465
T _{Outlet} (°F)	529	519-535	494	485-492
ΔT (°F)	39	28-40	34	23-35
H ₂ Conc (cc/kg)	25-35	10-35	25-35	11-20
O ₂ Conc (ppm)	<0.05	Nil	<0.05	Nil
pH	9.5-10.5	9.5-10.88	9.5-10.5	9.5-10.1
Crud (ppm)	<3.0	<0.02 (At start)	<3.0	<3.0

TABLE B-2. LOOP RADIOCHEMICAL DATA
BEFORE AND AFTER ACTIVITY BURSTS

Date	Cs ¹³⁸ (dpm/ml $\times 10^{-6}$)	Kr ⁸⁸ (dpm/ml $\times 10^{-6}$)	Gross $\beta - \gamma$ (cpm/ml $\times 10^{-6}$)
X-3-m Test Startup on May 6, 1960			
Periods Before and After First Activity Burst			
10-31-60	7.42	13.3	1.21
11-3-60	6.65	11.3	0.98
11-9-60	5.61	10.0	1.19
11-11-60	52.4	281.0	6.15
11-12-60	6.23	82.2	--
11-14-60	53.2	193.0	2.37
11-17-60	43.9	94.9	3.68
11-21-60*	--	--	--
11-26-60	3.72	4.20	1.48
11-29-60	2.98	4.67	0.73

*Experiment was removed, examined, reassembled, and returned for further irradiation after removal of failed fuel Elements M31 and M52.

Periods Before and After Second Activity Burst			
12-7-60	2.88	4.04	1.11
12-9-60	3.24	4.68	0.81
12-13-60	2.57	4.03	1.26
12-15-60	4.52	4.85	1.14
12-27-60	3.76	7.19	1.31
1-5-61	6.07	6.91	1.30
1-9-61	3.00	5.46	1.30
1-18-61	111.0	37.0	--
1-23-61**	--	--	--
1-26-61	4.85	5.58	--
2-1-61	4.49	6.09	3.00
2-3-61	3.05	6.67	1.62
2-8-61	2.75	5.93	1.57

**Experiment was removed, examined, reassembled, and returned for further irradiation after removal of failed fuel Element X6N12.

TABLE B-2 (Cont)

<u>Date</u>	<u>Cs¹³⁸ (dpm/ml x 10⁻⁶)</u>	<u>Kr⁸⁸ (dpm/ml x 10⁻⁶)</u>	<u>Gross β - γ (cpm/ml x 10⁻⁶)</u>
Periods Before and After Third Activity Burst			
2-14-61	3.07	5.88	1.67
2-16-61	3.06	6.94	--
2-17-61	--	--	--
2-20-61*	--	--	--
3-3-61	5.25	11.9	1.68
3-7-61	3.91	10.8	1.79
3-9-61	2.98	6.90	1.36
3-14-61	3.90	8.78	1.38

*Experiment was removed, examined, reassembled, and returned for further irradiation after removal of failed fuel Element X1B32.

Periods Before and After Fourth Activity Burst			
3-24-61	3.90	18.3	1.98
3-28-61	6.43	10.8	3.92
3-30-61	5.98	18.1	4.20
4-7-61	9.96	27.5	4.99
4-13-61	8.56	9.30	1.10
4-16-61	--	--	70.0
4-17-61**	121.0	269.0	98.8
4-27-61	2.40	2.10	0.87
5-3-61	1.69	2.12	0.60

**Experiment was removed, examined, reassembled, and returned for further irradiation after removal of failed fuel Elements X7N11 and M72.

TABLE B-2 (Cont)

Date	Cs ¹³⁸ (dpm/ml $\times 10^{-6}$)	Kr ⁸⁸ (dpm/ml $\times 10^{-6}$)	DN Monitor (cpm $\times 10^{-6}$)	γ Monitor (cpm $\times 10^{-3}$)	Gross $\beta - \gamma$ (cpm/ml $\times 10^{-6}$)
Periods Before and After Fifth Activity Burst					
11-1-61	1.15	1.56	--	--	0.37
11-6-61	--	--	--	--	--
11-7-61	1.19	1.78	1.5	1.5	0.43
11-8-61	1.25	--	--	--	--
11-15-61	1.13	1.66	1.3	1.4	0.35
X-3-n Test Startup on November 20, 1961					
11-27-61	0.90	0.95	1.2	7.0	0.24
11-28-61	--	--	--	--	0.25
12-4-61	--	--	1.8	7.0	0.24
12-12-61	--	--	1.8	7.0	0.20
12-15-61	--	--	2.1	6.6	0.09
12-21-61	--	--	0.8	6.5	0.22
1-3-62	0.62	0.78	--	--	--
1-9-62	--	--	0.8	5.6	0.15
1-17-62	--	--	0.8	5.6	0.21
1-18-62	9.71	25.5	7.2	42.0	0.84
1-22-62	--	--	--	--	--
1-31-62	--	--	5.4	8.4	0.28
2-5-62	0.93	1.06	5.1	8.5	0.27
2-9-62	--	--	5.1	8.5	0.32
2-13-62	--	--	4.8	8.3	0.17

*Experiment was removed, examined, reassembled, and returned for further irradiation after removal of failed fuel Element M22.

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