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FINAL REPORT ON THE FIRST FUEL ROD FAILURE
TRANSIENT TEST OF A ZIRCALOY-CLAD
FUEL ROD CLUSTER IN TREAT

R. A. Lorenz
D. C. Hobson
G. W. Parker

OAK RIDGE NATIONAL LABORATORY

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UNION CARBIDE CORPORATION

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REACTOR CHEMISTRY DIVISION

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MARCH 1971

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Oak Ridge, Tennessee
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R. A. Lorenz, D. O. Hobson* and G. W. Parker

ABSTRACT

The first fuel rod failure experiment in Transient Reactor Test Facility (TREAT) was performed with a seven-rod bundle of 27 in. long Zircaloy-clad UO_2 fuel rods in flowing steam atmosphere. A water reactor loss-of-coolant accident was simulated by operating TREAT reactor at constant power for 20 sec so that fission heat in the UO_2 pellets caused the Zircaloy cladding temperature to rise $72^\circ\text{F}/\text{sec}$ to a maximum of approximately 1800°F . The fuel rods were initially pressurized with helium between 115 and 215 psia (77°F) to simulate accumulated fission gas.

The Zircaloy cladding swelled and ruptured resulting in 48% blockage of the bundle coolant channel area at the location of maximum swelling. The average rod maximum circumferential swelling was 36%. Calculations related the hoop stress and ultimate strength at the onset of rapid expansion. The ideal gas law was used to calculate the rate of cladding expansion from measured rod temperature and internal pressure. Metallographic examination revealed ductile ruptures and significant oxygen pickup. Zirconium-steam reaction was 0.2%.

The center rod was preirradiated to 650 Mwd/MT in the Materials Testing Reactor (MTR) to build up an inventory of fission products and to determine irradiation effects on fuel rod failure characteristics. No irradiation effect was seen from this low-level irradiation on the swelling and rupture characteristics. A form of fuel rod failure propagation apparently occurred when chips of fragmented UO_2 from the rupture hole in the center irradiated rod contacted one of the outer rods resulting in stress concentrations and rupture of the outer rod at the point of contact.

The release of gaseous fission products ^{133}Xe and ^{85}Kr from the irradiated center rod was in the 0.1% to 0.2% range. The release of volatile fission products

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^{131}I and ^{137}Cs was similar indicating negligible retention on the inner Zircaloy cladding surfaces. Approximately 6.7% of the ^{131}I released from the center rod was in a chemically unreactive form, probably CH_3I .

1. INTRODUCTION

With modern light-water power reactors, one of the most serious postulated accidents is the loss-of-coolant accident (LOCA) in which coolant water is lost through a break in the primary piping system. Emergency core cooling systems (ECCS) have been designed and installed in these reactors to provide emergency cooling in the unlikely occurrence of such a postulated accident.¹ Zircaloy cladding of the fuel rods would undergo a severe temperature transient during a loss-of-coolant accident as a result of redistribution of heat stored in the UO_2 fuel during the short time interval between blow-down of the cooling water and complete application of the emergency cooling water. Many of the fuel rods would bow, swell, and rupture before the emergency cooling performed its function. Continuous heating of the Zircaloy cladding from fission product decay heat and from the reaction of Zircaloy with steam at high temperature would result in embrittlement of the cladding and gross damage to the core accompanied by the release of large amounts of fission products if the emergency coolant could not adequately cool the core.

A fuel rod failure study program was initiated at Oak Ridge National Laboratory in July 1968 as part of the ORNL Nuclear Safety Program^{2,3,4,5} in order to determine the characteristics and extent of fuel rod failure under loss-of-coolant accident conditions and to determine the effect of failure on emergency cooling effectiveness. The program

at ORNL includes transient (rapid heating) burst tests of single and clustered Zircaloy tubes in inert atmosphere, heat transfer and cladding behavior during the blowdown phase of the LOCA, high temperature rupture tests of irradiated fuel rods in steam atmosphere, and rupture tests of clusters of fuel rods in steam atmosphere in the Transient Reactor Test Facility (TREAT) reactor.

This report covers the first fuel rod failure experiment performed in TREAT. The experiment test assembly consisted of a seven-rod cluster of 27-in.-long Zircaloy-clad UO_2 fuel rods exposed to a flowing steam atmosphere in simulation of steam flow conditions immediately following the blowdown portion of the LOCA. The center rod contained fission products from a preliminary irradiation to a fuel burnup of 650 Mwd/MT to test the effect of irradiation on rupture characteristics as well as to provide a fission product inventory for fission product release measurements. The rods were prepressurized with helium which simulated fission gas accumulated after high burnup. The fuel rod failure test was performed in TREAT on September 12, 1969, by operating the reactor at steady power so that fission heat in the UO_2 pellets raised the cladding temperature in duplication of loss-of-coolant temperature behavior immediately following coolant blowdown.

By using fissioning in the UO_2 pellets as the heat source, the heat transfer conditions between pellet and cladding are the same as expected in the LOCA. The TREAT fuel rod failure experiments are therefore considered to be proof-tests of data and behavior models derived in the other tube-burst and fuel rod rupture research tasks.

2. DESCRIPTION OF EQUIPMENT — TREAT FUEL ROD FAILURE EXPERIMENT FRF-1

2.1 General Description

A photograph of the in-reactor components of the equipment is shown in Fig. 2.1. Fuel rod cladding was a nominal 0.564 in. diameter and the rods were located on an equilateral triangle spacing 0.75 in. apart. The rods occupied 51% of the cross-sectional area in the triangular lattice and 46% of the area within the 2-3/16 in. I.D. Zircaloy sleeve surrounding the rod bundle. Steam flowed up through the rod bundle at a rate of 11 l/min (STP) along with 1.8 l/min (STP) helium at a pressure of about 19 psia. The steam-helium mixture carried fission products from the ruptured rods through the filter packs where most of the aerosol particles and iodine were collected. The entire steam system was preheated electrically to 130°C to prevent condensation of steam. The primary vessel was well insulated to prevent overheating of the reactor fuel when the experiment fuel rod bundle was heated to a much higher temperature.

The flow system is shown in Fig. 2.2. Constant heat input of 440 watts to the steam generator provided 9 g/min steam flow to the bottom of the rod bundle. Helium at 1.8 l/min (STP) was mixed with the steam in the generator, and the mixture passed through a filter pack upon leaving the primary vessel. Chemically reactive compounds of the fission product iodine deposited on silver-plated surfaces and aerosol particles were collected on three fiberglass-asbestos filters in series. A high-efficiency silver-plated diffusion coil behind the filters trapped reactive iodine desorbed from particles on the filters. The steam was condensed and the helium carried remaining volatile fission products through a warm iodine-impregnated charcoal trap for collection of CH_3I and through liquid-nitrogen-cooled charcoal trap for

PHOTO 93731A

Fig. 2.1. In-Reactor Components of TREAT Fuel Rod Failure Experiment.

Fig. 2.2. Schematic Flow Diagram of TREAT Fuel Rod Failure Experiment.

collection of xenon and krypton. Effluent helium was monitored by rotameter and wet test meter. Hydrogen formed by reaction of steam with zirconium appeared as effluent gas flow greater than the controlled helium flow rate. A second identical fission product collection system was used after the first 90 sec in order to determine what fission products were slowly released after the initial rupture. Back-purging of Filter Pack No. 2 with helium prevented accidental contamination while Fission Product Collection Unit-1 was in use.

2.2 Fuel Rod Construction

Details of fuel rod construction are given in Table 2.1. The four rods with Zircaloy-2 cladding were surplus rods of commercial manufacture built to Dresden-I dimensions. The rods were used "as-received" so that precise internal dimensions were not obtained. Nominal dimensions were: cladding, 0.564 in. dia and 0.033 in. wall thickness, and UO_2 dia 0.492 in. The three rods with Zircaloy-4 cladding were assembled at ORNL using cladding of recent commercial manufacture. All rods had a combined fuel and plenum length of 26-7/8 in. Selected pellets from extra "Dresden-I" rods were used for fuel. The pellets averaged 0.62 in. long and had an O/U ratio of 2.001. The immersion density at 25°C in water averaged 10.34 (94.4% of theoretical), enrichment was 1.51%, and impurities were less than 100 ppm for each element identified. All cladding was inspected ultrasonically for flaws using a 0.002 x 0.002 x 0.5 in. groove as a standard. No defects were detected in these rods although most of the other rods from the Dresden-I size batch showed defects. The plenums of the three Zircaloy-4 rods each contained a spring wound from 1/16 in. dia Zircaloy wire. The spring deflection was 0.12 in./in.-lb, but the rods were assembled without compression on the springs. A 3/8 in. long Zircaloy cylinder with a 1/16 x 1/16 in. groove for free gas passage was located between the spring and first pellet.

Table 2.1. Fuel Rod Characteristics

ROD IDENTIFICATION:	CENTER	4-1	4-2	R	I	H	L
<u>Cladding Material:</u>	<u>Zr-4</u>	<u>Zr-4</u>	<u>Zr-4</u>	<u>Zr-2</u>	<u>Zr-2</u>	<u>Zr-2</u>	<u>Zr-2</u>
Cladding, O.D., in.	0.5637	0.5638	0.5638	0.568	0.570	0.564	0.566
Cladding, I.D., in.	0.4986	0.4989	0.4989	0.503	0.503	0.502	0.502
Wall Thickness, in.	0.033	0.033	0.033	0.032	0.033	0.031	0.032
Pellet Dia., in.	0.488/0.489	0.496/0.497	0.496	~0.492	~0.492	~0.492	~0.492
Gross Wt., g	965	1023	975	1074	1038	1096	1094
UO ₂ Wt., g	766	820	767	884	848	906	905
Plenum Length, in.	2-5/8	1-1/8	3-5/16	0	0	0	0
Plenum Volume, cm ³	7.55	3.23	9.51	0	0	0	0
Clad Gap and Pellet Gap Voids, cm ³	3.54	2.74	1.45	4.01	7.18	2.5	2.9
Pressure Cell and Tubing, cm ³	0	0	0	0	0	~3.0	~3.0
Total Gas Space, cm ³	11.09	5.97	10.96	4.01	7.18	5.50	5.92
Pressure, psia He at 77°F	215	215	215	215	115	133	142
Helium in Rod, cm ³ (STP)	149	80	147	54	52	46	39

A 1/8 in. dia Zircaloy tube with 1/16 in. dia hole was welded to the top of each rod for pressurizing. The rods were held at 255°F and evacuated overnight for drying and outgassing. The void volume was measured by expanding helium into the rod from a known volume. Pressure was increased to the desired level and the tube was pinched from the outside onto a 1/16 in. dia gold wire located in the tube. The pinch-seal was checked for leakage and the tube then cut and seal welded. A final leak check was made with a mass spectrometer helium leak detector. After assembly, the Zircaloy-4 rods were autoclaved for 2 days in 1500 psi steam at 750°F. The helium fill pressure range of 115 to 215 psia (77°F) was based on estimates of fission gas pressure in a BWR calculated by the D' (empirical) method.⁶ This calculation showed that most of the rods would contain pressure below this range, but that most of the volatile fission products released from UO₂ into the rod void spaces would be contained in rods with pressures above this range.

The differences in rod construction were made to see if any one variable strongly influenced the characteristics of clad swelling and rupture. Two cladding alloys were used, pellet to clad gap ranged from 0.002 to 0.010 in. on the diameter, total void volume ranged from 4.0 to 11.1 cm³, and internal pressure ranged from 115 to 215 psia (77°F). Rods H and L were each connected to a strain gauge pressure transducer for continuous monitoring of internal pressure. Both rods had two Pt vs Pt-Rh thermocouples made of 30 gauge wire spot-welded to the cladding, but two of these thermocouples, one on each rod, were damaged after initial assembly when alterations were made to allow increased linear expansion.

2.3 Fuel Rod Suspension

Rods H and L, which were connected to pressure cells, were hung from the top support spider and tightened to the spider so that rotation or other movement at that location

would be highly restrained. These rods and all others had 1/32 in. clearance on the dia with the bottom spider spacer. The other rods rested on a bottom support plate without any restraint on rotation. The top end of these rods had 1/64 in. clearance on the diameter in holes in the upper support spider. All rods had 5/16 in. available for linear expansion. The rods were not clipped or supported other than at the ends.

2.4 Center Rod Irradiation

The center rod was placed in a special irradiation capsule and irradiated as experiment ORNL-58-2 in the MTR from June 11 to June 25, 1969, for 13.125 full power days. The peak linear power heat rating for the irradiation was 15.2 kw/ft, and the peak-to-average ratio was approximately 1.15. Peak burnup was 650 Mwd/MT. Magnesium alloy spacers between the rod and outer stainless steel container provided the proper temperature drop between the fuel rod cladding and the relatively cool MTR cooling water. At the peak power location the UO_2 surface temperature was calculated to be 970°F and the centerline temperature 3450°F. After the irradiation the rod was neutron radiographed and gamma scanned with a lithium-drifted germanium scintillation crystal and a 0.005 in. slit-width collimator. The neutron radiograph showed that a central void did not form in the UO_2 pellets. An axial void would be expected from grain growth and sintering with sufficient time at temperatures above 3450°F. The gamma scan showed uniform fission product concentrations at pellet centers and at pellet interfaces. Previous experience with a group of special capsules showed a correlation between high heat rating, central void formation and the presence of radiation peaks at pellet interfaces caused by migration of fission products.^{7,8} After gamma scanning, the rod was removed from its irradiation capsule and installed in the center of the TREAT experiment

bundle in the TAN (Test Area North) hot cells at the National Reactor Testing Station.

3. EXPERIMENTAL PROCEDURE

The experiment was installed in TREAT, the entire flow system was tested with 30 psig helium for leakage, flow control valves were tested and the steam safety shutoff system was tested. Two calibration transients were performed at low reactor energy output to confirm the calculated rise in cladding temperature for a given reactor energy release. The first calibration transient was performed with the experiment evacuated and initially at room temperature to minimize heat loss. The second transient was performed with steam flow and the experiment preheated. Temperature response to the three transients is summarized in Table 3.2. The increase in primary vessel temperature resulted from reactor gamma heating.

The loss-of-coolant transient proceeded according to the following schedule:

-14 min	Steam flow started (11 l/min STP), helium flow: 1.8 l/min (STP) through primary vessel and 1.8 l/min (STP) additional purge through Filter Pack-2.
0 min	Start of TREAT transient.
8 sec	Reactor power steady at 30 Mw.
28 sec	Reactor power decreasing from 30 Mw. Maximum measured cladding temperature of 1770°F reached. Primary vessel pressure increase from 7.6 to 10.5 psig caused mainly by helium released from rods.
28.4 sec	Rod H ruptured at ~172 psia.
28.7 sec	Rod L ruptured at ~162 psia.

Table 3.2. Temperature Response to Reactor Transients

		TREAT Integrated Power Mwsec	Duration of Reactor Power sec	TC-7 (Rod H) °F	TC-9 (Rod L) °F	TC-3 (Primary Vessel) °F
Calibration Transient #1:	Start	0	0	94.6	95.4	84
	End	38.9	2	229.5	233.6	86
Calibration Transient #2:	Start	0	0	365	365	327
	End	195.3	10	918	903	343
Loss-of-Coolant Transient:	Start	0	0	361	356	331
	End	556	20	1771	1679	388

90 sec Flow changed to Fission Product Collection Unit-2.
 10.5 min Heat to steam generator turned off.
 16.25 min Flow changed back to Unit-1.
 18 min Steam generator bypassed.
 30 min Flow stopped.

4. EXPERIMENTAL RESULTS

4.1 Pressure, Temperature, and Flow Rates

Measured reactor power, cladding temperature and rod internal pressures are shown in Fig. 4.3. TC-7 was 9-5/16 in. below the top of rod H at a location receiving 99% of the peak flux. TC-9 was 6-1/4 in. below the top of rod L where the flux was about 94% of the peak. The cladding temperature rate of rise averaged 72°F/sec. Peaking of the pressure curves at 23 sec (1560°F) probably corresponds to the beginning of significant volume increase by swelling. The pressure increase in these two rods should have been slower than in other rods because of the unheated gas volume in the tubing and pressure cell (see Sections 4.8 and 4.9).

System pressures and flow rate are shown in Fig. 4.4. Primary vessel pressure was obtained from gauges located outside of the reactor so that response was slow. A sensitive pressure cell located close to the primary vessel failed during mockup testing and could not be replaced. Therefore the individual pressure pulses from rupturing rods were not obtained. The large rise in pressure between 25 and 30 sec indicates that most of the rods failed within this time span. The decrease in primary vessel pressure at 90 sec when flow was changed is a result of less flow restriction in Unit-2. The gradual rise in pressure after 150 sec is undoubtedly due to a buildup of flow restriction in one of the freeze traps. Increased flow rate of steam leaving the primary

Fig. 4.3. Fuel Rod Temperatures and Pressures in TREAT Experiment FRF-1.

Fig. 4.4. System Pressure and Flow Rate During TREAT Fuel Rod Failure Experiment FRF-1.

vessel between 25 and 35 sec results from higher pressure in the primary vessel. The slightly lower flow rate between 36 and 60 sec may represent a conversion of steam to hydrogen. This reaction is on a 1-to-1 volume ratio, but the rotameter does not respond significantly to hydrogen because of its low density.

4.2 Metal-Water Reaction

The wet test meter at the outlet of the flow system measured combined helium and hydrogen flow. The effluent gas volume curve in Fig. 4.4 was obtained from wet test meter readings with the constant helium flow subtracted. No correction was made for the effect of temperature increase or pressure changes in the primary vessel, or for helium released from the rods. The volume of helium released from the rods was 738 cm^3 at 12.5 psia and 77°F , the ambient conditions of the wet test meter. Temperature and pressure changes in the system account for about 1.3 liters of helium effluent between times 0 and 120 sec. Therefore we estimate the volume of hydrogen generated by metal-water reaction to be 1.2 ± 0.6 liters (STP). This is equivalent to about 0.2% metal-water reaction based on total cladding volume.

4.3 General Description of the Bundle as Removed from the Capsule

The experiment was dismantled in the TAN facility hot cells. The bolts holding the top end-cap were unscrewed and the capsule was upended to slide the rods part way out of the capsule. Photographs were taken of the rods as they were removed to illustrate the relative positions of the ruptures and the overall appearance of the rupture region. The fields of view of the various photographs are illustrated in Fig. 4.5. The photographs, Figs. 4.6 through 4.15, show that the ruptures occurred on the inside of the rod bundle. Figures 4.6 and 4.7, taken from approximately the same positions, show

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Fig. 4.5 Diagram of the Cross Section of the TREAT Bundle Illustrating the Directions (a through j) From Which Photographs Were Taken. The diagram represents the bottom of the rod cluster, but it is also the view of the upended bundle as seen from the top of the hot cell.

Fig. 4.6 Periscope View (a) of Rods R, 4-2 and L (left to right). The rupture in Rod R is visible as well as the cladding fragment from one of the ruptures.

Fig. 4.7 View (b). Same as Fig. 7, Except for Slight Rotation of the Bundle to Allow Better Examination of the Fragment.

Fig. 4.8 View (c). Overall View of the Bundle After Approximately 75% Withdrawal From the Capsule. The rupture region is visible near the center of the photograph.

Fig. 4.9 View (d). Photograph of Rods 4-2, L, I and H (left to right). The close approach of Rods L and I is apparent.

Fig. 4.10 View (e). Photograph of Rods L, I and H.

Fig. 4.11 View (f). Photograph of Rods I, H, 4-1 and R. The center rod is just visible.

Fig. 4.12 View (g). Overall View of the Bundle
Approximately 180° from Fig. 4.9.

Fig. 4.13 Views (h and i). Photographs of Rods H, 4-1 and R. A portion of the rupture bulge in the center rod is visible. Two photographs have been mounted to show the entire length of the rupture zone of the bundle.

Fig. 4.14 View (j). Photograph of Rods 4-1, R, 4-2 and L Showing the Rupture in Rod R Straddling Rod 4-2.

Fig. 4.15 Photograph of Rods H, Center and R After Removal of Rod 4-1. Rod R has been rotated approximately 120° from its burst position.

a small fragment of cladding caught between rods 4-2 and L. Figure 4.8 is an overall view of the partially removed bundle taken at the same angle as Fig. 4.7. The small cladding fragment seen in the two previous figures is no longer seen and presumably fell to the hot cell floor during handling of the assembly. Figure 4.9 shows the closest approach, except for point contact between a rupture and an adjacent rod, of any of the rods along any appreciable length of the bundle. Figures 4.10 and 4.11 are two other views taken around the bundle. The rods were pulled out of the bottom spacer as the bundle was removed from the primary vessel, so the rods may have been in more intimate contact than is shown in the photographs.

Figure 4.12 is an overall view of the bundle taken at a point opposite Fig. 4.8. Figure 4.13 is a composite showing the bulge in the center rod and the entire length of the rupture zone of the bundle. Figure 4.14 shows the rupture in rod R straddling rod 4-2. Figure 4.15 shows the center rod after removal of rod 4-1. The relative positions of the rods in this photograph were shifted when 4-1 was removed.

4.4 Fuel Rod Failure Characteristics and Propagation

The bundle was further dismantled and the rods were photographed head-on and sideways to the ruptures. A montage is shown in Figs. 4.16 and 4.17. The horizontal background lines represent one inch intervals referenced from the top shoulders of the rods. The numbers associated with the rods are maximum diametral expansions as viewed in the photographs. All ruptures occurred within two inches axial distance along the bundle.

The center rod contacted every other rod at some point with sufficient pressure to cause metal deformation. Most points of contact can be seen in Figs. 4.16 and 4.17. Figure 4.18 is a top view of the bundle plan showing direction of rupture and direction of rod-to-rod contact. The center rod contacted rod 4-1 at 10.7 in. from the top shoulder, rod R

Fig. 4.16 A Montage Showing Head-On Views of the Ruptures in the Rods.

Fig. 4.17 Side View of Ruptures.

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Fig. 4.18 Top View of Bundle Plan Showing Direction of Rupture and Rod-to-Rod Contact.

at 11.6 in., rod 4-2 at 11.0 in., rod L at 11.6 in., rod I at 10.2 in. and rod H at 12.0 in. Other rod contacts which caused deformation occurred between rods 4-1 and H at 10.7 in., rods 4-2 and L at 10.9 in., and rods 4-2 and R at 11.6 in. The median location for rod-to-rod contact was 11.0 in., precisely the location found for maximum coolant channel blockage.

The contact between the center rod and rod H occurred almost directly at the points of rupture. The metal on the right side of the rod H rupture as seen in Fig. 4.16 was depressed more than 1/16 in. by a piece of material approximately 1/16 in. square. We believe that a chip of UO_2 from the ruptured center rod caused the depression.

Calculations based on hoop stress show that the center rod should have ruptured several seconds before rod H. The pellets in the center irradiated rod were cracked into small pieces and the pieces may have settled inside the rod as the cladding swelled, thus making a somewhat concentrated heat source. The direct effect on rod H was probably the spilling out of UO_2 chips that heated and caused local stress concentrations where the expanding cladding of rod H pressed against the UO_2 pieces. This form of fuel rod failure propagation would not occur in out-of-reactor fuel rod failure tests and apparently would not occur in the TREAT experiments with unirradiated rods, since the unirradiated pellets remained intact.

The rods were examined for bowing by placing them against a large, vertical sheet of graph paper so that the maximum bowing was perpendicular to the axis of view through the window of the hot cell. Figure 4.19 illustrates the rods in position against the graph paper. Bowing occurred mainly away from the rupture side. The maximum bowing occurred in rod H and we calculated that the rod axis was displaced approximately 0.15 in. from its original centerline. Since the average radial clearance between the rods and the Zircaloy sleeve was approximately 0.060 in., it is probable that some of the rods started bowing, hit the sleeve and then rotated to allow further bowing to take place.

PHOTO 98124

Fig. 4.19 Photograph of the Rods Taken Against Graph Paper to Show Maximum Bowing. The bowing can be seen easily by viewing the photograph from the bottom at a low angle.

4.5 Metallographic Examination of Rod 4-2

The Zircaloy-4 cladding from rod 4-2 of FRF-1 was sectioned for metallographic examination and photographed in both the as-polished and etched conditions. Three immediate observations were made: (1) the rupture was ductile, (2) there were heavy oxide films on both the inner and outer surfaces, and (3) there was a large temperature gradient around the circumference of the tube.

Figure 4.20 illustrates a transverse cross section taken at the rupture, or at a position corresponding to 1-3/8 in. on the scale of Fig. 4.21. An unetched cross section taken from an undistorted region of the tube was mounted inside the rupture cross section (Fig. 4.20) to illustrate the swelling that occurred. The darker areas counterclockwise from the rupture to approximately 12 o'clock and clockwise from the rupture to approximately 8 o'clock were in the $\alpha + \beta$ and/or the β regions during the experiment.

Figure 4.22 (point A of Fig. 4.20) shows one end of the rupture region and the large amount of wall thinning associated with the ductile behavior of the material. The apparent difference in oxide film thicknesses is real and can be explained by the fact that the outer surface of the tubing was exposed to the steam atmosphere first and then, following rupture, both the inner and outer surfaces were exposed.

The maximum oxide thickness was found on the lip of the rupture and was approximately 0.0017 in. Because of the large temperature gradients, both along and around the tube, the oxide thicknesses varied quite markedly. The outside surface oxide thickness 180° from the rupture was only 0.0003 in. The oxide thickness seems to be proportional to temperature.

Fig. 4.20 Transverse Cross Section of Rod 4-2 Taken Through Rupture. An unetched cross section (Y-98626) from an undistorted region of the tube is mounted inside the rupture cross section. Arrows indicate regions discussed in text. 5.25X

Y-97668

Fig. 4.21 Rupture Region of Rod 4-2

Fig. 4.22 Point A of Fig. 4.20 Showing Oxide Films and Ductile Rupture. 50X

Examination of the microstructure in the cross section at the rupture showed a temperature gradient of approximately 200 to 300°F in the maximum temperatures reached by the cladding. This estimate is based on the evidence of $\alpha + \beta$ and β phases having been formed during the temperature transient. Figure 4.21 shows a structure that transformed from 100% β back to α during the slow cooling that took place after rupture. The minimum equilibrium $\alpha + \beta$ -to- β transformation temperature is approximately 1780°F. Oxygen contamination would tend to raise this temperature. Figure 4.23 (point B of Fig. 4.20) shows a structure that formed upon slow cooling from the high $\alpha + \beta$ region. This photograph was taken approximately 0.5 in. along the circumference from the rupture. Traces of the large prior- β grains may be seen. The maximum temperature of this region would have been approximately 1700 to 1750°F. Figure 4.24 (point C of Fig. 4.20) shows a region that was subjected to a maximum temperature corresponding to the α -to- $\alpha + \beta$ transformation. At equilibrium this temperature is approximately 1500°F, and this should be considered the minimum transformation temperature for this region. Point C is approximately 1.7 in. from and almost diametrically across from the rupture. Figure 4.25 (point D of Fig. 4.20) shows the microstructure of the coolest region, and judging from grain size and lack of indications of former β phase the maximum temperature was probably between 1450 and 1550°F.

Hardnesses varied considerably both along the tube length and around the circumference. Longitudinal hardnesses, based on distance from the rupture, are shown in Fig. 4.26. Tube 4-2 had a circumferential wall thickness variation along its length, with the thin wall region lying in a straight line down the tube length. The rupture occurred in this thin wall region. Figure 4.26 shows curves for hardnesses measured in the centers of the thinnest portions of the wall and in the thickest part of the bulge or at the lips of the rupture.

Fig. 4.23 Point B of Fig. 4.20 Showing Structure Formed by Cooling from the High β Portion of the $\alpha + \beta$ Phase. Large prior β grains are outlined. 50X

Fig. 4.24 Point C of Fig. 4.20 Showing Structure Formed by Cooling from a Temperature Near the α -to- $\alpha + \beta$ Transformation Temperature. Both α and prior $\alpha + \beta$ can be seen. 50X

Fig. 4.25 Point D of Fig. 4.20 Showing Structure in Coolest Region of Rupture Cross Section, Probably All α Phase. 50X.

Fig. 4.26 Hardness of Zircaloy-4 Tubing, Rod 4-2.

The high hardness values in the bulge were due to one main circumstance: the relative decrease in wall thickness in the bulge, leading to decreased oxygen diffusion distances, allowed increased oxygen penetration into the mid-wall layer. This hardening effect probably occurred during expansion and after tube rupture, during the long cool-down period.

Figure 4.27 is a schematic representation of the section cut through the center of the rupture. Hardness values are shown, corresponding to adjacent points on the drawing, that illustrate the large amount of hardening near the rupture. A photograph of this section, in Fig. 4.20, shows the microstructural variations caused by uneven temperature distribution in the test bundle. The higher temperatures near the rupture area would also aid oxygen diffusion and hardening. Except at the lips of the rupture, there was negligible formation of oxygen-stabilized α phase.

Room temperature ductility can be estimated from the hardness measurements based on correlations observed when standard three-point bend tests (MAB-192-N) were made at cross-head travel speed of 1 in./min with 0.040 in. thick Zircaloy-4 sheet containing dissolved oxygen.⁹ We estimate that most of the cladding would have bend ductility of 90° (DPH hardness approximately 180) and that the lips of the rupture would have bend ductility of 12° or less (DPH hardness above 400).

Fig. 4.27 Hardness Measurements in Cross-Section of Rod 4-2 at Rupture Location, Viewed Toward Top of Rod.

4.6 Swelling Measurements

Diameters of outer rods were measured at increments along the length using micrometers in line with the ruptures and at 90° . The average diameter increases are shown in Fig. 4.28 for the Zircaloy-4 rods and in Fig. 4.29 for the Zircaloy-2 rods. There was no apparent difference in swelling between the two Zircaloy alloys. Swelling helped to provide an open path connecting the plenums (rods 4-1 and 4-2) with the rupture zone.

The temperatures shown were calculated from an assumed initial axial preheat temperature distribution, the increase in temperature recorded by the two thermocouples, and axial distribution of fission heat based on the measured flux profile. The flux profile was determined by gamma scans of the outer rods and is shown in Fig. 4.30. The solid line is an unperturbed curve expected if a TREAT fuel element occupied the experiment location. Significant flux depression occurred in the lower half of the experiment from a heavy molybdenum liner placed there to contain melted Zircaloy cladding that might result from an accidental full-power TREAT transient. Cladding that faced the center rod was probably hotter than the listed temperatures while that which faced the outer Zircaloy sleeve was probably cooler.

4.7 Coolant Channel Blockage

The average diameter was measured along the length of each rod using micrometers on the outer rods and photographs of the center rod. Individual coolant channel size could not be measured directly, so we subtracted the total rod cross sectional area from the total sleeve area to obtain the remaining coolant area. The resulting average coolant channel blockage is shown in Fig. 4.31 as a function of axial distance. The greatest average blockage was 48% and was near the center of the rupture zone. The sleeve area

Fig. 4.28 Swelling of Zircaloy-4-Clad Fuel Rods in TREAT Experiment FFF-1.

Fig. 4.29 Swelling of Zircaloy-2 Clad Fuel Rods in TREAT Experiment FRF-1.

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Fig. 4.30 Flux Profile in TREAT Experiment FRF-1 (points) and Expected in Unperturbed Experiments (solid line).

Fig. 4.31 Coolant Channel Blockage for BWR Rod Spacing.

was approximately equal to the total area occupied by seven rods and coolant channels in a modern BWR, so the calculated average blockage is the same for BWR rod spacing. Since the outer rods were all exposed to the cool Zircaloy sleeve, the expansion and coolant channel blockage data is most representative of rods in a reactor that are adjacent to cool surfaces such as bundle shrouds (BWR) or control rod thimbles (PWR).

4.8 Rate of Expansion of Cladding

Knowledge of the rate of swelling should help elucidate the mechanism of cladding swelling and rupture. To calculate the fuel rod volume increase, use was made of the ideal gas law,

$$nR = P \left(\frac{V_1}{T_1} + \frac{V_2}{T_2} + \frac{V_3}{T_3} \right) ,$$

where nR is constant until rupture, P is the rod pressure, V_1 is the pressure cell volume (1.5 cm^3), V_2 is the volume of connective tubing coiled inside the primary vessel (1.5 cm^3), and V_3 is the fuel rod void volume (2.5 cm^3 initially for rod H and 2.92 cm^3 initially for rod L). The pressure cell temperature T_1 was observed to rise linearly from 285 to 312°F during the first 28 sec, whereas T_2 , the temperature of the tubing volume, was estimated to rise from 361 to 514°F during the first 28 sec, with the rate of rise being proportional to the quantity $(T_3 - T_2)$. The temperature of the fuel rod void volume, T_3 , was assumed to be equal to that determined by TC-7, the thermocouple on rod H. Further refinement, such as providing for the fuel rod axial temperature gradient, was not warranted because of limited quantity and accuracy of the available data.

The results of the calculations are given in Fig. 4.32. The rods expanded rapidly for 2 or 3 sec before rupturing, and approximately 80% of the volume increase occurred in

Fig. 4.32 Internal Pressure, Temperature, and Volume Expansion of Rods in Experiment FEF-1 in TREAT.

this period. Actual volume increases calculated from micrometer measurements for rods H and L were 9.2 and 11.6 cm³, compared with calculated volumes of 7.2 and 10.2 cm³, respectively.

Calculations of cladding swelling under loss-of-coolant conditions have also been made at Battelle Memorial Institute.¹⁰ For BWR conditions similar to those of the ORNL experiment, they predict the beginning of rapid swelling at approximately 1400°F and the attainment of strains in excess of 30% at 2 to 3 sec later. ORNL's FRF-1 experiment indicated this to be true, except that rapid swelling of rods H and L began at approximately 1600°F.

4.9 Prediction of Rupture Temperature for Experiment FRF-1

Use of the ultimate strength and simple hoop stress formula ($2st = pd$, where s is stress, t is wall thickness, p is pressure drop across tube wall, and d is tube diameter) to predict the tube rupture temperature can be tested with some legitimacy by determining when the maximum internal pressure is reached and the cladding begins to expand. For the average between rods H and L, $s = pd/2t = (278-20)(0.500)/(2 \times 0.0315) = 2050$ psi. This corresponds to the ultimate tensile strength of Zircaloy-4 at 1650°F.¹¹ At this temperature the cladding was expanding rapidly. Because of the thermocouple location and the radial temperature gradient, the temperature toward the center of the bundle was higher than the measured temperature.

The ideal gas law and ultimate tensile strength can be used further to calculate the maximum possible pressure in the fuel rods. The fuel rods were all of different construction, so the maximum pressures, as well as pressures at time of rupture, were somewhat different. For the average wall thickness and tube diameter, the ratio of hoop stress to

pressure drop across the wall is approximately 8. The internal pressure required to reach the ultimate strength is shown in Fig. 4.32. According to these data the measured pressure in rods H and L exceeded that necessary to reach the ultimate strength. This is undoubtedly a result of inaccuracies in the pressure and temperature measurements, combined with inadequacy of the calculational model. It is believed that the effect of strain rate on ultimate strength is very important. The referenced ultimate strength was measured at a strain rate of approximately 0.06/min, which is very slow compared with the experimental swelling rate. Workers at ORNL are including the effect of strain rate in their analytical model of Zircaloy swelling.¹³ Indications are that a factor of 10 increase in strain rate could double the apparent cladding strength.

The onset of rapid expansion may be easier to predict than the conditions at rupture. By using the ideal gas law to calculate internal pressure with no expansion, the stress in rods H and L reached the referenced ultimate strength as rapid swelling began. For example, assuming no swelling, at 24.0 sec the internal pressure in rod L would have been 305 psi, a pressure sufficient to stress the cladding to the low-strain-rate ultimate at 1610°F.

Similar calculations were performed for other rods, and the corresponding pressures, temperatures, and times are presented in Table 4.1. The effect of the differences in rod construction may be seen; a large plenum (rod 4-2) reduced the maximum pressure compared with a small plenum (rod R). The calculated maximum pressures are only approximate, since slow swelling would have reduced the maximum pressures, especially in rods R and I, which had no plenums. The time predicted for onset of rapid expansion is indicated for each rod along the temperature curve in Fig. 4.32.

Table 4.1 Conditions at Beginning of Swelling and at Rupture of Zircaloy-Clad Fuel Rods

	Rod R	Rod 4-1	Center Rod	Rod 4-2	Rod I	Rod L	Rod H
Initial pressure, psia at 77°F	215	215	215	215	115	142	133
Beginning of swelling							
Estimated time, sec	19.8	21.2	21.6	21.7	22.0	24.0	24.5
Estimated maximum pressure, psia	690	490	455	450	420	305	285
Measured pressure, psia						285	272
Estimated temperature, °F	1370	1470	1490	1500	1520	1610	1640
Measured heatup rate, °F/sec	65	65	65	65	60	45	45
Conditions at rupture							
Measured volume increase, cm ³	10.9	10.1	~11	10.1	9.6	11.0	9.2
Estimated pressure, psia	215	250	290	205	190	150	155
Measured pressure, psia						162	172
Conditions after rupture							
Maximum circumferential swelling, %	36	35	40	42	35	36	26
Hole size, mm ²	150	15	30	23	6	7	4

The actual amount of swelling was measured with micro-meters after disassembly of the cluster. The volume increases were calculated from these measurements and are also listed in Table 4.1. By knowing the final volume and assuming a 4-sec delay between calculated maximum pressure and rupture, it was possible to estimate the internal pressure at time of rupture. These pressures are also given in Table 4.1.

4.10 Fission Product Release

The release of fission products when fuel rods rupture during a loss-of-coolant accident is dependent mainly on the previous time and temperature of fuel irradiation. High temperature and long irradiation time increase the diffusion of fission products from the UO_2 pellets into the clad gap and plenum where they are available for rapid release if the cladding ruptures during a loss-of-coolant accident. Long half-life fission products will accumulate in the clad gap and plenum, but the amounts of short half-life fission products in the clad gap and plenum are dependent mainly on the fuel temperature during the latest reactor operating time corresponding to one or two fission product isotope half-lives.

Details of the center rod irradiation in the MTR were presented in section 2.4. During the loss-of-coolant accident simulation in TREAT volatile and gaseous fission products were released rapidly from the clad gap and plenum of the ruptured center rod, and only small additional amounts were released from the UO_2 . This amount, released when UO_2 is heated above previous operating temperature, has been called the "heating burst." Both heating bursts and cooling bursts were measured when UO_2 pellets were heated and cooled during post-irradiation annealing experiments to measure fission product release.¹³

Fission product release in the TREAT experiment is summarized in Table 4.2 and complete details are given in Table 4.3. Table 4.2 shows that the release of the volatile fission products ^{131}I and ^{137}Cs was similar to the release of gaseous fission products ^{133}Xe and ^{85}Kr . The behavior of ^{129}Te was similar to that of ^{137}Cs .

Fission products with low volatility (^{89}Sr , ^{95}Zr , ^{103}Ru , ^{140}Ba , and ^{141}Ce) did not, within the precision of our data, fractionate from each other or therefore from the UO_2 . The maximum cladding temperature attained was about 1800°F . Our chemical analysis was for uranium rather than UO_2 , and all seven rods released some uranium (or UO_2) whereas only the center rod contained significant fission products. Based on the low-volatility fission products, mentioned above, the amount of uranium released from the center rod was about half of the total found. In other words, the center irradiated rod released six times as much uranium (or UO_2) as each un-irradiated rod. This greater release of uranium may be a result of increased surface area provided by cracking and recoil effects in the irradiated rod. Total uranium found in the filter packs was 11 mg.

The rate of release of various fission products can be determined by comparing the amounts found in units 1 and 2. Almost all the ^{133}Xe (99.5%) was found in unit 1, proof that flushing of the primary vessel was completed within 62 sec following rod rupture. More than 98% of the "solids" were collected in unit 1, as opposed to about 70% of the ^{131}I , ^{129}Te , and ^{137}Cs . This demonstrates a significant slow or delayed release of iodine, tellurium, and cesium. Feuerstein investigated the system of I_2 in Zircaloy and reports both prompt and slow release of ZrI_4 at investigated temperatures from 1100 to 1475°F .¹⁴

The ^{131}I carried out of the primary vessel into the fission product collection system has been characterized by

Table 4.2 Fission Products Released in TREAT Fuel Rod Failure Experiment FRF-1

Location	Material Found in Each Location (% of Total in Center Irradiated Rod) ^a						
	¹³¹ I	¹³⁷ Cs	¹²⁹ Te	Fission Products with Low Volatility ^b	U	¹³³ Xe	⁸⁵ Kr
Primary vessel ^c	0.054	0.046					
Filter Pack: Deposited by diffusion	0.076	0.0004	0.0001				
Filter Pack: Deposited with particles	0.046	0.010	0.015	0.0009	0.0002		
Condensate	0.0001						
Heated Charcoal	0.013						
Total Release	0.189	0.056				0.14	0.094

^aFRF-1 irradiated 13.1 full-power days to 645 Mwd/MT peak burnup at 15.2 kw/foot peak linear power, peak/average flux 1.15, total fissions 9.8×10^{21} .

^bMedian of ⁸⁹Sr, ⁹⁵Zr, ¹⁰³Ru, ¹⁴⁰Ba and ¹⁴¹Ce.

^cPrimary vessel leached with 0.5 N NH₄OH. Only soluble materials reported.

Table 4.3 Complete Tabulation of Fission Product and Uranium Distribution Found in TREAT Loss-of-Coolant Experiment No. P22-1

Location	Material Found in Each Location. 10 ⁻⁴ %											
	¹³¹ I	¹²⁹ Te	¹³⁷ Cs	¹⁰³ Ru	¹⁴⁰ Ba	⁸⁹ Sr	¹⁴¹ Ce	⁹⁵ Zr	U	¹³³ Xe	^{131m} Xe	⁸⁵ Kr
Primary Vessel	543	25 (1)	457	0.34 (1)			0.061 (1)	0.16 (1)				
<u>Filter Pack, Unit-1:</u>												
Loose Particles	73	7.5	6.6	3.34	3.26	3.78	8.51	4.07	0.74			
Housing	68	6.3	7.2	7.354	0.382	0.475	1.15	0.463	0.1445			
Flow diffuser	144	6.5	4.4	0.148	0.161	0.188	0.101	0.188	0.0747			
Diffusion coil #1	228	0.36*	1.95	0.220	0.186		0.152	0.238				
Diffusion coil #2	94	0.91*	1.39	0.129	0.117		0.102	0.145				
Diffusion coil #3	60	0.36*	1.41	0.071	0.052		0.047	0.750				
HEPA filter #1	169	79	51.3	3.02	5.05	3.40	6.80	3.83	1.106			
HEPA filter #2	0.95											
HEPA filter #3	0.99											
Diffusion coil #4	17	0.02*	0.0060	0.0016	0.7034		0.0009	0.0018				
<u>Filter Pack, Unit-2:</u>												
Housing	68	7.8	2.24	0.11	0.120	0.136	0.082	0.141	0.032			
Flow diffuser	156	11.7	8.0	0.0164	0.011*	0.0195	0.011	0.0179	0.0067*			
Diffusion coil #1	70	0.06	1.54	0.0001*	0.0018*		0.001*	0.0004*				
Diffusion coil #2	20	0.21	0.97	0.0037	0.0022		0.0016*	0.0004*				
Diffusion coil #3	21	0.04*	1.08	0.0011	0.0009*		0.0006*	0.0003*				
HEPA filter #1	18.0	26.4	15.6	0.0087	0.0166	0.0193	0.0127	0.0178	0.0048*			
HEPA filter #2	0.1											
HEPA filter #3	0.3											
Diffusion coil #4	12	0.03*	0.020	0.0002	0.00003		0.0001*	0.00007*				
<u>Collection Unit-1:</u>												
Condensate	0.46			0.0017								
Heated Charcoal Trap	87											
Rare Gas Trap No. 1A										1379	2813	716
Rare Gas Trap No. 1B										24	48	210
<u>Collection Unit-2:</u>												
Condensate	0.43											
Heated Charcoal Trap	38											
Rare Gas Trap No. 2A										7.1	19	10
Rare Gas Trap No. 2B												
<u>Totals:</u>												
Init-1	943	101	74	7.28	9.20	7.85	16.9	9.01	2.07	1403	2861	930
Unit-2	404	46	29	0.143	0.153	0.175	0.109	0.179	0.044	7.1	19	10
Units 1 and 2	1347	147	103	7.42	9.35	8.03	17.0	9.19	2.11	1410	2880	940
Units 1 and 2 and primary vessel	1890		560									

(1) These elements were not completely soluble in the 0.5 N NH₄OH leach.

*Plus or minus 100%.

three forms, based on its distribution in filter packs, condensate, and heated charcoal traps. One classification is particulate iodine, that which is on the surface or within released solid particles such as UO_2 , U_3O_8 , or ZrO_2 . The second form is chemically reactive molecular forms such as I_2 , HI , or HOI that diffuse rapidly and tend to deposit quickly on surfaces such as the silver-plated diffusion coils. The third form is unreactive iodine, such as the organic iodide CH_3I .

The fission product collection systems were designed to facilitate this classification. Figure 4.33 is a photograph of a filter pack for collection of fission products carried out of the primary vessel. The flow entered the cone and passed through silver-plated brass-ribbon diffusion coils for collection of diffusing iodine species. Only three coils were used ahead of the filter papers in FRF-1 because of space limitations. Three fiber-glass-asbestos HEPA filter papers (Cambridge type 1G) were used to collect particles, and a closely wound high-efficiency silver-plated diffusion coil collected iodine vapor desorbed from particles on the filters or other surfaces.

One problem with samplers of this type is that some particles stick to surfaces intended for rapidly diffusing vapor forms of iodine, and these surfaces are not always perfect sinks for the molecular forms (I_2 , HI , and HOI). Also, particulate iodine may sorb and desorb after sampling, and iodine vapor sorbed on filter media is frequently troublesome.

In the following analysis, particulate iodine throughout each filter pack was based on the amount of solids (median of ^{103}Ru , ^{140}Ba , ^{89}Sr , ^{141}Ce , and ^{95}Zr) at a given location and by the ratio of ^{131}I to solids found on the first filter paper. The ^{131}I /solids ratio on the first filter of filter pack 1 was $169 \times 10^{-4}\% / 4.4 \times 10^{-4}\% = 38$.

Fig. 4.33 Components of the Filter Packs for Fission Product Collection Are Silver-Plated "Diffusion Coils," Three Fiber-Glass-Asbestos HEPA Filters, and a High-Efficiency Backup Diffusion Coil.

The amount of particulate ^{131}I on the backup diffusion coil, for example, was obtained by multiplying the solids concentration by 38, or $(0.001 \times 10^{-4}\%) \times 38 = 0.038 \times 10^{-4}\%$. The remaining ^{131}I was presumed to have been deposited by diffusing iodine species. Table 4.4 is a complete classification of ^{131}I by this procedure. The ^{131}I /solids ratio in filter pack 2 was $18 \times 10^{-4}\% / 0.017 \times 10^{-4}\% = 1050$, significantly higher than that found in filter pack 1. All the ^{131}I in the condensate and heated charcoal traps was of the unreactive organic iodide form. This is confirmed by the solubility or partition coefficient between the helium and condensate at 32°F .

The relatively large amount of unreactive organic iodides is of special interest. Almost 7% of the ^{131}I carried out of the primary vessel into the fission product collection system was apparently chemically unreactive and, based on the experience of others, was probably CH_3I . The partition coefficient of the unreactive ^{131}I species may be estimated from the rate of helium flow and condensate formation in the condenser tubes at 32°F . In 1 min, 9 ml of water was condensed from 3600 cm^3 of helium. Defining partition coefficient as the ratio of concentration in the liquid to concentration in the gas,

$$p = \frac{0.88/9}{135/3600} = 2.8 \quad .$$

For CH_3I we would expect a partition coefficient of about 8.5, and for I_2 , at low concentrations, about 10,000. Low concentration may contribute to the relatively large proportion of unreactive iodine. Approximately 2.0 mg of iodine fission products, mostly ^{129}I , were in the center rod at the time of the TREAT test. Approximately $3.6 \mu\text{g}$ were collected in the primary vessel and fission product collection systems. During the first minute after rod rupture we estimate that

Table 4.4 Distribution of ^{131}I in Experiment FRF-1^a
(Values are in percent $\times 10^{-4}$)

	Unit 1	Unit 2	Units 1 and 2
Particulate iodine			
On large loose particles	73	0	73
On housing	18	68	86
On flow diffusing screens	7	18	25
On diffusion coil 1	8	1	9
On diffusion coil 2	4	1	5
On diffusion coil 3	2	1	3
On HEPA filter 1	169	18	137
On backup diffusion coil	0.07	0.9	1
Total solids-associated	281	108	289
Diffusing iodine			
On housing	50	0	50
On flow diffusing screens	137	138	275
On diffusion coil 1	220	63	289
On diffusion coil 2	90	19	109
On diffusion coil 3	58	20	78
On HEPA filter 1	0	0	0
On HEPA filter 2	1	0.1	1
On HEPA filter 3	0.9	0.3	1
On backup diffusion coil	17	12	29
Total diffusing iodine	574	258	832
Organic and other unreactive forms			
In filter pack	0	0	0
In condensate	0.46	0.43	0.89
On warm iodine-impregnated charcoal	87	38	125
Total "organic"	89	38	126

^aBased on total inventory of center rod.

2.5 μg were released and exposed to about 39 liters of steam and helium in the primary vessel at 1015°F, an average concentration of 64 $\mu\text{g}/\text{m}^3$. The concentration would be about twice this in the filter pack at 265°F.

The higher concentration of iodine on solids in unit 2 is duplicated by ^{129}Te and ^{137}Cs . We might speculate that oxidation of UO_2 to U_3O_8 formed solid particles that carried more iodine, tellurium, and cesium. Oxidation of UO_2 by steam at 1740°F would not release fission products with low volatility, based on results with steam oxidation of uranium.¹⁵ Particles from the first filter paper of filter pack 1 were examined with the electron microscope and with selected area transmission electron diffraction. Figure 4.34 is a photograph of this filter paper. UO_2 was found, along with a metallic oxide of the spinel type with lattice spacings similar to Fe_3O_4 . Several lines, moderately strong for U_3O_8 , were observed in the UO_2 pattern. No indication of any zirconium compound was found. Much of the material in the electron micrographs appeared to be amorphous by electron diffraction. Crystallite size for the material giving diffraction patterns was in the region 0.1 to 1.0 μ .

In spite of the different iodine concentration on particles, the relative amounts of particulate, diffusing, and unreactive iodine were the same in both filter packs. The diffusing iodine was not distributed according to calculations for molecular iodine diffusing to perfect adsorbers. Calculations gave a decontamination factor of 25 for each diffusion coil and a factor of 1000 for the high-efficiency backup diffusion coil.¹⁶ Fouled silver surfaces could have caused poor retention of molecular forms of iodine.

Fig. 4.34 First Filter of Fission Product
Collection Unit-1, TREAT Experiment FRF-1.

5. DISCUSSION

5.1 Operating Conditions

The operating conditions were selected to be representative of loss-of-coolant accident conditions for a BWR, particularly for the short time interval near the end of coolant blowdown and before arrival of emergency coolant. This is the time period during which the cladding temperature will heat up quickly and many fuel rods will swell and rupture. In the TREAT experiment we used a steam flow rate of 11 liters/min (STP) or 1.2 lb/hr. Based on total cladding area, this corresponds to steam flow of 34,800 lb/hr in the Browns Ferry reactor, 33,400 lb/hr in a 2255-Mw(t) BWR, and 27,400 lb/hr in a 2760-Mw(t) PWR. In the loss-of-coolant accident, steam flow decreases slowly after the main blowdown, and the flow rates above correspond to a time after blowdown of 7 sec for the PWR and 50 sec for BWR's, as estimated at Argonne National Laboratory in computer studies of typical loss-of-coolant accidents.^{17,18}

The initial preheat temperature of 360°F was lower than cladding temperature in a reactor, but no fuel-cladding interaction or clad swelling was expected during the initial temperature increase above 360°F, especially at the relatively slow rate of 72°F/sec. For example, at 72°F/sec heatup rate the cladding temperature did not lag much behind the UO₂ pellet temperature so that thermal expansion of the UO₂ could not have created unusual stresses or strains in the Zircaloy cladding.

The linear heatup rate of 72°F/sec was not selected to match a particular hypothesized loss-of-coolant accident. The maximum cladding temperature of approximately 1800°F was higher than anticipated for some hypothesized accidents with efficient emergency cooling, but it is considerably lower

than might occur in other hypothesized accidents with degraded emergency cooling. Temperature higher than that required to swell and rupture the cladding would have little effect on fission product release but would be expected to increase the amount of Zircaloy-steam reaction and to increase the embrittlement of the Zircaloy by oxygen pick-up.

5.2 Swelling and Rupture Characteristics

No effect of irradiation on the swelling or rupture characteristics of the Zircaloy cladding on the center irradiated rod was observed. This rod received only a 13 day irradiation in the MTR which was not expected to measurably affect either the strength or ductility of the cladding.

There was no difference in behavior of the Zircaloy-4 tubing obtained from a recent commercial batch and Zircaloy-2 tubing manufactured a number of years ago in connection with Dresden-I tubing development or production.

Oxidation by steam had no effect on swelling or ruptures in that the ruptures were all ductile. The ruptures occurred at temperatures between 1600 and 1800°F where zirconium-steam reaction rates are relatively low. Some loss of room temperature ductility was observed during the metallographic examination of rod 4-2 (Section 4.5).

The fuel rods were constructed with different plenum sizes. Many reactor fuel rods are built with plenums that occupy approximately 10% of the fuel rod length. From Table 2.1 we see that the center rod and rod 4-2 fit this description and rods H and L with the low-temperature pressure cells probably behaved like fuel rods with large plenums. Visual observations did not reveal any effect of plenum size on swelling or rupture. However in Section 4.9 and Table 4.1 we showed that plenum volume does affect the temperature and pressure conditions at the beginning of swelling and at rupture. Since swelling of a full length reactor fuel rod might

occur only within a two-foot long hot zone, the experiment rods with the largest plenums were probably the most realistically proportioned for loss-of-coolant swelling and rupture studies.

The pellets in rods 4-1 and 4-2 (Table 2.1) were selected for piston-like fit in the tubing. The pellets fit as close as possible without requiring force during loading of the tubing. We wanted to see if the close fit would tend to isolate the plenums from the region of swelling and rupture. This effect was not observed, possibly because of the short distance between plenum and rupture and possibly because swelling along the entire length of the rod increased the pellet-to-clad gap significantly.

The internal pressure had a definite effect on the rupture characteristics. The low-pressure rods ruptured with smaller holes compared with larger razor-edged openings in the high-pressure rods. This is probably mainly an effect of temperature on Zircaloy properties at the temperature of rupture. Hobson performed a series of tube-burst tests in inert atmosphere and observed that ruptures which occurred in the Zircaloy β phase (above 1780°C) were small-holed with an orange-peel textured surface on the expanded metal.¹⁹ Ruptures below this temperature in the $(\alpha + \beta)$ two-phase region were larger-holed with sharp flared edges and smooth metal surfaces.

Hobson found that minimum circumferential expansion occurred when Zircaloy was in the two-phase $(\alpha + \beta)$ region. With tubing similar to that used in the TREAT experiment this corresponded to a maximum pressure differential across the tubing of 400 to 600 psi. In Table 4.1 calculated maximum pressure differentials of 265 to 670 psi are shown. This is evidence that the circumferential expansion observed in TREAT experiment FRF-1 may be close to the minimum that can be expected and that either lower or higher initial

pressures might result in greater expansion. The circumferential expansions listed in Table 4.1 agree with Hobson's data except that the low-pressure rods in TREAT expanded less. The large radial temperature gradient across the bundle, the steam atmosphere, and unknown wall thickness variations could have contributed to the lower expansion.

5.3 Coolant Channel Blockage and Fuel Rod Failure Propagation

The observed circumferential expansion (36% average maximum) and calculated coolant channel blockage (48% maximum) is most representative of locations in a power reactor where a cool surface such as a bundle shroud or control rod thimble is adjacent to the fuel rods. Waddell performed fuel rod failure tests in inert atmosphere with 13-rod bundles of the same type tubing used in the TREAT experiments.²⁰ His pressurized tubes were 24 in. long with 16 in. heated by internal quartz-encapsulated tungsten heaters. He found slightly less coolant channel blockage when his peak rod pressure was about 440 psig (Test 4) and a coolant channel blockage curve almost identical to TREAT FRF-1 (Fig. 4.29) when his peak rod pressure was about 220 psig in Test 5.

In both types of tests the failures were close together. In TREAT the ruptures were within a 2 in. axial length. In Waddell's 13-rod tests the locations of maximum swelling usually occurred within 3 or 4 in. This is probably an indication of high sensitivity to temperature. In a power reactor similar regions of concentrated failure might be expected in a loss-of-coolant accident since high-power-density regions frequently occur in bundles following the gradual withdrawal of nearby control rods.

A form of fuel rod failure propagation was observed in experiment FRF-1 where ruptures in the center irradiated rod and in outer rod H were found to be superimposed as discussed

in Section 4.4. The hypothesized mechanism for failure propagation is that the pellets in the center rod were cracked into small pieces from the previous MTR irradiation and the pieces may have settled inside the rod during the TREAT experiment as the cladding swelled, thus making a concentrated heat source. The center rod ruptured with an opening large enough to spill out chips of UO_2 against rod H so that additional heating and mechanical stress concentrations occurred where the expanding cladding of rod H pressed against the UO_2 pieces.

This form of fuel rod failure propagation would not occur in out-of-reactor fuel rod failure tests and apparently would not occur in TREAT experiments with unirradiated rods since the unirradiated pellets remained intact. The ideal test of this failure propagation mechanism would be with a larger bundle of all-irradiated rods.

5.4 Fission Product Release

Fission product release mechanisms and data were discussed in Section 4.10. Some comments on the behavior of iodine might be in order. It has been hoped that the fractional release of ^{131}I from fuel rods undergoing a loss-of-coolant accident might be considerably less than the fractional release of fission gases such as ^{133}Xe and ^{85}Kr . Optimistically the ^{131}I release in a loss-of-coolant accident might be a factor of 20 less than the fission gases because of lower iodine "diffusion" from UO_2 during normal reactor operation and from iodine retention on the inner Zircaloy cladding surfaces during the loss-of-coolant accident. No significant difference in fractional release was observed in experiment FRF-1 between the volatile fission products ^{131}I and ^{137}Cs and the fission gases ^{133}Xe and ^{85}Kr .

Another observation of potential importance to reactor safety is the relatively high proportion of chemically

unreactive ^{131}I released during the TREAT experiment. Approximately 6.7% of the released ^{131}I apparently was in the form CH_3I based on the experience of others with similar fission product collection systems and based on the observed partition coefficient (solubility in water). It is possible that the particular combination of temperature, concentration, atmosphere (steam-hydrogen), and container material used in the loss-of-coolant experiment was conducive to the formation of organic iodides.

The general level of release of volatile and gaseous fission products was lower than expected. The irradiation conditions for the center rod (Section 2.4) were intended to be similar to those of a medium to high power density rod of a modern power reactor where fission gas release would be between 1% and 10% for a short irradiation. The D' (empirical) method was used to estimate the fission gas release from UO_2 to the fuel rod plenum and void spaces during the MTR irradiation.²¹ This calculational method is based on fission gas release from a series of capsule (short rod) irradiations performed by AECL (Atomic Energy of Canada Limited) and correlates fission gas release with linear power and irradiation time, but does not include corrections for UO_2 temperature variances caused by cladding temperatures or clad gap conductivities different from that of the reference capsules.

The center rod peak linear power was 15.2 kw/ft based on a radiochemical burnup analysis. The peak to average ratio was 1.15 based on an MTR flux profile so that the average rod linear power was 13.2 kw/ft. Based on previous calculations we expect that fission gas release from a rod with axial power distribution will behave like a uniform-power capsule operating at the average of the rod peak and rod average linear power ratings. Therefore the center rod would be comparable to a capsule operating at 14.2 kw/ft. The reference capsules contained UO_2 of lower density and

thermal conductivity so that fission gas release from the center rod would be the same as that from a reference capsule operating at $0.98 (14.2) = 13.9$ kw/ft or 456 w/cm linear power. The D' (empirical) method was then used to obtain the calculated fission product release results shown in Table 5.1. We use an empirical diffusion parameter for iodine four times that of xenon and krypton. The empirical diffusion parameter D'_1 should not be confused with the conventional diffusion coefficient D' .

The measured releases are a factor of 25 or 30 below the calculated releases. Retention on the inner surface of the Zircaloy cladding could affect iodine, but not xenon or krypton. We believe that the reason for the low fission product release may be inferred from Figure 5.1, a metallographic cross-section of the center rod taken below the rupture and close to the location of peak power during the MTR irradiation. The peak linear power of 15.2 kw/ft, obtained from radiochemical analysis, gives calculated UO_2 surface and centerline temperatures of 520°C (970°F) and 1900°C (3452°F), respectively. The limited grain growth shown in Fig. 5.1 is typical of temperatures lower than 1900°C and is also typical of structures exhibiting low fission product release. The elongated grains near the center are associated with the healing of cracks during the MTR irradiation and should not be confused with columnar grains usually formed at higher temperatures. The short heating cycle to 1800°F during the TREAT experiment should not have affected the UO_2 grain structure.

Since the limited grain growth correlates with low fission product release and with maximum temperatures below the calculated 1900°C (3452°F), we believe that the calculated UO_2 temperatures are probably too high. Therefore the observed release of volatile and gaseous fission products is typical of the short irradiation of a low power rod rather

**Table 5.1 Comparison of Calculated Release from UO₂
and Measured Release from Center Rod**

	Fission Product Isotope		
	¹³³ Xe	¹³¹ I	⁸⁵ Kr
Empirical Diffusion Parameter D ₁ ', sec ⁻¹	2x10 ⁻¹⁰	8x10 ⁻¹⁰	2x10 ⁻¹⁰
Calculated Fractional Release at Secular Equilibrium	0.034	0.085	-
Equilibrium Correction Factor	0.82	0.68	-
Calculated Release from UO ₂ (%)	2.8	5.8	3.4
Measured Release from Center Rod (%)	0.14	0.19	0.094
Ratio, Calculated UO ₂ Release/ Measured Rod Release	20	30	36

PHOTO-101329

Fig. 5.1 Metallographic Cross-Section of the Center Rod Near the Location of Peak Power in the MTR.

than a medium or high power rod.

We would like to have examined the inner surface of the Zircaloy cladding on the center rod for direct measurement of retained ^{131}I and ^{129}I . This phase of the fission product examination was not included in the TREAT fuel rod failure program, so we remain uncertain about the magnitude of iodine retention by the Zircaloy cladding. Further experimentation is needed, preferably using ^{129}I to confirm the behavior of ^{131}I , before we can confidently compare the release behavior of iodine and the fission gases.

6. CONCLUSIONS

The TREAT fuel rod failure experiment FRF-1 was conducted under the most realistic loss-of-coolant accident conditions of any experiment to date. Fission heating in the UO_2 pellets provided precise duplication of the heat transfer conditions between pellets and cladding expected in a loss-of-coolant accident. We cannot form definite conclusions from this single experiment, but much of the data is directly relevant to reactor safety and siting and should be explored further using similar experiments.

Of particular significance, we found that the ruptures and swollen areas were close together, within a 2 in. axial distance. This indicates high sensitivity to temperature and low sensitivity to random defects such as wall thickness or strength variations. The magnitude of maximum swelling and the rupture characteristics were in general agreement with tube burst tests performed in inert atmosphere in spite of a large radial temperature gradient across the TREAT fuel rod bundle.

The fractional releases of volatile fission products ^{131}I and ^{137}Cs were similar to those of the fission gases ^{133}Xe and ^{85}Kr , an indication that credit should not be assumed for slower diffusional release of ^{131}I from UO_2 during normal reactor operation and for retention of ^{131}I on the surfaces of Zircaloy cladding undergoing loss-of-coolant accident conditions. Total fission product release was low, apparently because of the irradiation conditions in the MTR, but the large proportion of ^{131}I released in TREAT in a chemically unreactive form (probably CH_3I) indicates that the particular combination of temperature, atmosphere, containment material and concentration might be conducive to the formation of organic iodides.

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