INTERIM REPORT

IRRADIATION TESTING OF CERAMIC FUELS

For Period
October 1, 1963 through November 30, 1964

Prepared by:

G. Zuromsky
J. J. Koziol
W. P. Chernock

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1

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Argonne, Illinois 60440

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Mr. E. J. Kreh
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IRRADIATION TESTING OF CERAMIC FUELS

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The following personnel contributed to the project during this period:

Project Engineer	G. Zuromsky, CEND
Out-of-Pile Testing and Evaluation	G. Zuromsky, CEND J. J. Koziol, CEND
Irradiation Program	W. P. Chernock, CEND G. Zuromsky, CEND
Capsule Irradiations	E. O. Fromm, BMI J. H. Stang, BMI
Post-Irradiation Evaluations	W. A. Wagner, BMI V. W. Storhok, BMI J. E. Gates, BMI G. Zuromsky, CEND W. P. Chernock, CEND

I. INTRODUCTION

In previous work at CEND, the limits of composition were established for thermally stable, single-phase, face-centered cubic crystal structures in the ZrO_2 -rich and CeO_2 -rich regions of the UO_2 - ZrO_2 - CaO and UO_2 - CeO_2 systems. In addition, physical, thermal, and corrosion properties were determined. An irradiation program is in progress to demonstrate the high burnup capability and the inherent irradiation stability of single-phase, face-centered cubic solid solutions containing 4 and 10.2 m/o UO_2 in CaO -stabilized ZrO_2 and 7 and 17.5 m/o UO_2 in CeO_2 .

The irradiation testing of the first three instrumented capsules, each containing five stainless steel clad fuel samples, was initiated in the ETR and the MTR during November 1962, while the second three capsules of the series were inserted in the ETR during April 1963. Target burnups for each composition are 40, 55 and 70% fission of the U-235 atoms. Emphasis has been placed on maintaining proper temperature control by scheduling series of capsule moves to provide higher fluxes as burnups proceed. The first two capsules, 43-1 and 43-4, have been discharged and post-irradiation examination is in progress. Irradiation testing of the remaining four capsules is continuing in the ETR to burnups of 55 and 70% of the U-235 atoms.

Concurrent with these in-pile investigations, out-of-pile tests have been performed. Long time isothermal heat treatments of sintered pellets of the four fuel compositions have been initiated to simulate specific temperature zones of the in-pile fuel pellets. The purpose is to separate the long term irradiation effects from the thermal effects in the interpretation of

post-irradiation data. Thermal conductivity testing of the zirconia-base compositions has been completed at temperatures up to 2400°C. In addition, an indication of the temperature required for the onset of massive grain growth was evaluated as a possible aid in post-irradiation evaluations. Corrosion tests of bare pellets of the four fuel compositions are continuing in 360°C static degassed water and 625°C flowing steam. All pellets were evaluated every 90 days for weight change and general appearance.

This report summarizes progress on this program for the period October 1, 1963 through November 30, 1964. Although definitive conclusions concerning in-reactor fuel performance can not be made at this time, all pertinent data are presented. Another interim report will be prepared when post-irradiation analyses of samples from the first two irradiation capsules have been completed to provide complete post-irradiation analyses of fuels from these capsules.

II. SUMMARY

Two capsules, containing fuel samples of 10 and 22 wt% (4 and 10.2 m/o) UO_2 in CaO stabilized ZrO_2 and 10 and 25 wt% (7 and 17.7 m/o) UO_2 in CeO_2 have successfully completed irradiation testing in the ETR and MTR to fission burnups of about 40% of the U-235 atoms and at target temperatures just below melting. Irradiation of the remaining four capsules is continuing to target fission burnups of 55 and 70% of the U-235 atoms. Post-irradiation evaluations are now in progress on samples of the four fuel compositions irradiated in the first two capsules.

Although the correlation between temperature gradient data and exposure data from dosimeter wires were not good, both sets of data indicated that the target burnups were generally achieved. Fission gas release values ranged from 20% for the Z-10 (4 m/o UO_2) fuels to 75% for the C-25 (17.7 m/o UO_2) fuels, with intermediate values of 25% for the C-10 (7 m/o UO_2) fuels and 50% for the Z-22 (10.2 m/o UO_2) fuels. Although good correlation was obtained between volume fraction of grain growth and fission gas release, the reason for this good correlation is not understood, especially in light of the temperature history of the fuel which indicated temperature peaks at different times during the irradiation for different fuel samples.

Examinations of the microstructure of the irradiated samples indicated that the two ZrO_2 -base fuels had undergone more extensive changes than the CeO_2 -base fuels as evidenced by void formation and agglomeration, fission gas agglomeration, and development of large radial grains. There is some indication that the onset of grain growth in the CeO_2 -base samples occurred

at a temperature substantially below that of the ZrO_2 -base samples. All fuels exhibited a multiphase area at the edge of the grain growth region. It is postulated that this was a result of fission product migration. Formation of metallic particles was observed in all fuels. It is postulated that these particles are either U-Zr-Fs or U-Ce-Fs alloys. There was no metallographic indication of transformation of the original single-phase face-centered cubic solid solutions.

Measurements of fuel dimensions and densities are in progress. Mass spectrometric burnup analyses and X-ray diffraction investigations are yet to be performed. An interim report, containing evaluation of all data for samples in the first two capsules, will be issued when the present series of hot cell evaluations is completed.

Concurrent out-of-pile evaluations were conducted to provide information which would be useful in the post-irradiation evaluations. Long time phase stability studies indicated retention of the single-phase, face-centered cubic crystal structure after 250 to 300 days at temperatures of 1400 to 1475°C and 1380 to 1425°C for the zirconia- and ceria-base compositions. The metallic phase and the duplex structure (believed to be a result of fission product migration) observed in the irradiated fuels, were not detected, lending support to the contention that these structures are irradiation induced.

Thermal conductivity data for the zirconia-base fuel compositions were extended to 2200°C and indicated a gradual decrease in conductivity at temperatures up to 1500°C followed by an increase in conductivity at temperatures above 1500°C.

Unclad pellets of the four fuel compositions showed corrosion penetrations of less than 0.002 cm (0.001 inch) following 700 days exposure to 650°C flowing steam and 480 days in 360°C static degassed water.

III. THERMAL CONDUCTIVITY

A. Introduction

Out-of-pile tests were conducted on the ZrO_2 -base fuel samples to obtain thermal conductivity values at temperatures up to 2200°C . In addition, an attempt was made to obtain microstructure-temperature relationships. These data will be of importance in the post-irradiation examination of these fuels.

B. Test Apparatus

Techniques and apparatus developed at CEND⁽¹⁻⁴⁾, as part of a Joint United States Research and Development Program, have been utilized in this investigation of these ceramic fuels. The major features of the test equipment shown schematically in Figure 1, are discussed in detail below:

1. Test Element

The test element consisted of a column of ZrO_2 -base fuel pellets 1.17 cm (0.460 inch) diameter, clad in stainless steel and containing a tungsten-rhenium heater rod located axially through the pellet stack. The test element configuration is shown in Figure 2. The cladding was 1.270 cm (0.5 inch) OD stainless steel tubing with an 0.0508 cm (0.020 inch) thick wall. Thirteen 0.635 cm (0.250 inch) stainless steel standpipes were brazed to the cladding to provide optical paths to selected depths within the pellets for temperature measurements. Temperatures were measured optically at three radial locations within the ZrO_2 -base fuels as well as at the surface of the heater rod.

The fuel pellets were ground to OD and bore tolerances of

TEST APPARATUS
Fig. 1

— $\frac{1}{2}$ " TEST ELEMENT —
ARRANGEMENT OF COMPONENTS
Fig. 2

less than ± 0.003 cm (± 0.001 inch), which provided zero clearance between pellets and the cladding and the tungsten-rhenium heater at test temperatures. After assembly was completed, each test element was radiographed to ensure that all pellets were properly positioned within the cladding.

The pellets were prepared by cold compaction and sintering techniques, described previously⁽⁵⁾. Pellet densities ranged from 90 to 92 percent of theoretical.

2. Cooling Jacket

The cooling jacket (Figure 3) was a brass cylinder, flanged at both ends to permit easy insertion and removal of the test element. The row of 13 standpipes extended through the side wall of the jacket where they were sealed with O-rings. Both ends of the test element were also sealed with O-rings to the flanged ends of the cooling jacket. The annulus between the test element and the cooling jacket contained flowing deionized water for heat removal. This water was recirculated and purified in an ion exchange column, and cooled in a heat exchanger.

3. Test Chamber

The cooling jacket-test element assembly was contained within a stainless steel chamber approximately 30.48 cm (12 inches) long, 15.24 cm (6 inches) wide, 12.70 cm (5 inches) deep. The chamber cover, containing the viewing window, was removed for insertion and removal of the test element. Liberal use of O-rings permitted quick access into the chamber and provided adequate hermetic sealing. The leak rate of the chamber, blanked off from the pumps, was less than 0.5×10^{-3} Torr per minute. A vacuum level of about 10^{-5} Torr was attained within 30 minutes from a cold start.

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*13 STANDPIPE COOLING JACKET WITH
TEST ELEMENT INSERTED*

Power terminals, at both ends of the chamber, consisted of water-cooled copper bellows with a split copper clamp to provide quick coupling to the tungsten-rhenium heater. This design provided high current capacity while accommodating axial thermal expansion of the heater. The terminals were electrically insulated from the stainless steel chamber by plexiglass plates and hermetically sealed by O-rings.

All tests were conducted in inert gas at pressures slightly above atmospheric.

The atmosphere control system included a diffusion pump, with its associated cold trap and mechanical pump; a second mechanical pump for preliminary pumpdown; gas purification furnaces containing zirconium chips; and a pressure relief system to prevent pressure buildup in the test chamber. Appropriate high vacuum valves provided good flexibility of operation.

Fuel temperature measurements were made with a laboratory type optical pyrometer equipped with a prism at the objective end to permit sighting in a vertical direction. Reproducibility of temperature measurement from operator to operator was found to be within 2°C . Corrections for specimen emittance and optical path transmission were obtained by comparison of optical pyrometer readings with thermocouple readings. No evidence of sight glass degradation due to possible vapor deposition was observed. Cladding temperatures were measured with chromel-alumel thermocouples spot welded to the out surface of the cladding.

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C. Test Conditions

Pellet preparation, assembly, thermal cycling and examination procedures of the test elements are important functions in the overall test program. Caution must be exercised in the grinding and assembly operations to maintain the close tolerances required to minimize temperature drops at the heater-pellet and pellet-clad interfaces.

Three test elements were evaluated for thermal performance. Their designations and fuel compositions are summarized below:

<u>Element Designation</u>	<u>Fuel Composition*</u>
IRRAD-#1	Z-10
IRRAD-#2	Z-22
IRRAD-#3	Z-10

Descriptions of pellet preparation, element assembly, thermal cycling and examination procedures are included below.

1. Pellet Preparation

The outside diameter of the pellets was finish ground to provide a cold clearance of less than 0.003 cm (0.001 inch) with the stainless steel clad. The bore of the pellets was similarly ground to provide intimate pellet-heater rod contact during assembly. All pellets were end faced to a length of 0.635 cm (0.250 inch). The compositions and dimensions of the various components of the test elements are tabulated below.

<u>Component</u>	<u>Material</u>	<u>Dimensions (cm)</u>
Clad	304 Stainless steel	1.270 OD x 1.168 ID
Pellet	Z-10 or Z-22	1.266 OD x 0.231 ID
Heater	W-26 Re	0.229 OD

*Z-10: CaO stabilized ZrO_2 containing 10 wt% UO_2 .

Z-22: CaO stabilized ZrO_2 containing 22 wt% UO_2 .

Thirteen out of the 32 pellets in each element were slotted axially for temperature measurements (Figures 2 and 4). Initially, slots 0.038 cm (0.015 inch) in width were cut radially into the pellets to the desired depths. After measurements were made to confirm the depth of the slots, a second axial cut widened the slot to 0.102 cm (0.040 inch) to a depth of 0.254 cm (0.100 inch) from the pellet ID. This widened slot minimized obstruction of the optical path by pellet cracking during test. The depths of sight holes for various positions along the length of each test element are shown in Figure 4.

2. Element Assembly

The fuel pellet column was assembled by individually inserting pellets over the heater rod, which was positioned in the center of the clad. Each slotted pellet was positioned beneath its standpipe and then aligned by a shaped pin which was inserted through the standpipe and into the slot in the pellet. Retention of the alignment pins within the pellet slots during assembly prevented rotational displacement of the slotted pellets in relation to the standpipes. The assembled test element was radiographed to establish proper assembly of the pellet column within the clad. Prior to test, the assembly was vacuum dried at 125°C for 8 to 10 hours. The test element was inserted into the cooling jacket and the assembly installed into the test chamber for evacuation to a pressure of about 10^{-5} Torr. The system was backfilled to atmospheric pressure with inert gas and the test element outgassed further by increasing the temperature of the heater to about 300°C. Several cycles of evacuation followed by backfill of inert gas were performed to ensure complete removal of moisture prior to the start of the high temperature tests.

IRRAD No. 1

IRRAD No. 2

IRRAD No. 3

LEGEND

* = THIS SURFACE PREPARED FOR METALLOGRAPHY

SIGHT HOLES and POST-TEST SECTIONS
Fig. 4

3. Thermal Cycling

The test element was in readiness for thermal cycle tests upon attainment of a constant pressure of about 10^{-5} Torr within the test chamber. The thermal history of each test element is presented below.

a. Test Element IRRAD-#1

The temperature cycle history for element #1 is shown graphically in Figure 5. Following successful completion of the first cycle (heater rod temperature of about 1850°C for 2 hours), a longer time, higher temperature cycle was initiated at 1900°C . One end of the element formed a hot spot after 15 hours at temperature. Attempts to stabilize the temperature fluctuation by reductions in power were unsuccessful and burnout of the heater occurred near sight hole #12. The test was terminated and the element was sectioned for microstructural examination in accordance with the plan shown in Figure 4.

b. Test Element IRRAD-#2

Test element #2 was held at about 2400°C for 4-1/2 hours. A second cycle to 2400°C resulted in failure of the heater rod at sight hole #10 after about 1-1/2 hours at temperature, terminating the test. The sectioning plan for this element is also shown in Figure 4.

c. Test Element IRRAD-#3

Because of the failures of test elements #1 and #2 in the same general location, the axial temperature variation was reduced by installing diffusers on the inlet and outlet lines of the coolant jacket. This resulted in a somewhat more uniform coolant velocity along the length of the test element.

IRRAD No. 1
Z 10

IRRAD No. 2
Z 22

IRRAD No. 3
Z 10

CENTERLINE TEMPERATURE HISTORY
Fig. 5

Test element #3 was cycled with more success than elements #1 and #2. After a slow heatup it was maintained at 2100°C for 1 hour and then the temperature was decreased to 1000°C where it was held overnight. During the second cycle, the element temperature was increased to 2250°C. After about 3 hours at this temperature, a fluctuation in the power supply caused the equipment to trip out and the temperature to suddenly drop. The element was quickly brought to 1000°C where, again, it was held overnight. During the third cycle, the element was brought to 2400°C and held for 4-1/2 hours before being cooled slowly to room temperature.

It was found that the axial temperature variations (as a result of the change in the cooling jacket design) were much smaller than those for elements #1 and #2. In addition, the element had already completed three cycles at high temperature without failure. The purpose of the fourth cycle was to cause failure. After reaching 2500°C, temperatures could no longer be measured because of the closure of the sight hole by what is believed to be a vaporization-deposition of the fuel. The element failed shortly after the last temperature readings were made. The complete cycle history for element #3 is shown in Figure 5. After completion of the thermal cycling, the element was sectioned as shown in Figure 4 for metallographic examination.

D. Test Results

1. Thermal Performance

a. Thermal Conductivity

The thermal conductivities of the ZrO₂-base fuels were calculated on the basis of the following assumptions:

(1) The fuel pellets were in intimate contact with both the tungsten-rhenium heater rod and the cladding at the test temperatures.

(2) The heater rod-fuel pellet gap conductance was a constant value of $2 \text{ watts-cm}^{-2}\text{-}^{\circ}\text{C}^{-1}$.

(3) The contribution of cracks to the thermal resistance of the fuel was insignificant at operating temperatures.

(4) The fuel pellets were homogeneous, isotropic bodies. Actually, the pellets were continuous single phase structures with uniformly distributed porosity.

The temperature at the inside diameter of the fuel pellets was calculated from the average heater rod temperature, the geometry of the element, and the power required to maintain a given temperature within the element.

Values of thermal conductivity as a function of temperature were calculated by applying the integrated form of Fourier's law for heat transmission by conduction across a homogeneous material.

$$k = \frac{Q \ln \frac{R_2}{R_1}}{2\pi L (T_1 - T_2)} \quad \frac{\text{watts}}{\text{cm-}^{\circ}\text{C}}$$

where k = Thermal conductivity at a temperature $\frac{T_1 + T_2}{2}$

Q = Heat flux

R_1 and R_2 = Radial distances (cm)

L = Length of heat transfer surface (cm)

T_1 and T_2 = Temperatures at radial distances R_1 and R_2 ($^{\circ}\text{C}$)

The calculated values for thermal conductivity were corrected to 100% theoretical density by the simplified Loeb⁽⁶⁾ expression:

$$k_s = \frac{k_m}{1 - P_v} = k_m \frac{\rho_T}{\rho_m}$$

where k_s = Thermal conductivity of solid (100% dense) fuel

k_m = Measured thermal conductivity

P_v = Pore volume fraction

ρ_T = Theoretical density of fuel

ρ_m = Actual density of the fuel

Thermal conductivity data are presented in Figure 6.

Although test elements #1 and #3 were both Z-10 composition, results of element #3 were selected as representative of that fuel composition because of the early failure and erratic behavior of test element #1. The conductivity data obtained from the innermost radial segment ($R_1 - R_2$) of element #3 was totally unrealistic and erratic due to the deterioration of the heater rod and, therefore, not included in Figure 6. This, in effect, reduced the ΔT of the $R_1 - R_2$ segment and therefore increased the calculated k value for that radial segment. Additional metallographic observations of this region are reported in a later section.

The Z-10 data presented in Figure 6 resulted from the temperature observations at three radial positions. A high degree of correlation among the data points exists resulting in a rather narrow scatter band. The conductivity values for this composition remained within a relatively constant

Fig. 6

region between 0.0175 to 0.0225 watts-cm⁻¹-°C⁻¹ up to 1400°C. Above 1400°C the conductivity slowly increased and values at 1850°C were between 0.0200 to 0.0275 watts-cm⁻¹-°C⁻¹. The conductivity values for this composition remained within this region for three successive temperature cycles indicating no permanent influence of prior high temperature treatment on the conductivity.

The thermal conductivity values for the Z-22 composition also remained within a relatively constant region between 0.0150 to 0.0200 watts-cm⁻¹-°C⁻¹ up to about 1600°C. Above 1600°C a slow rise was observed to values between 0.0225 to 0.0275 watts-cm⁻¹-°C⁻¹ at 2200°C. These values are for one cycle only, since early failure of the test element prevented additional thermal conductivity determinations.

Published data ⁽⁷⁾ for UO₂ and CaO stabilized ZrO₂ indicate increased elevated temperature thermal conductivity. Similar results have been obtained on this program. In addition, results obtained by CEND for Phillips Petroleum Company as part of their Power Burst Program also exhibit increases in elevated temperature thermal conductivity, as shown in Figure 7.

The data, shown in Figure 7, indicate that the rate of increase in conductivity with increasing temperature appears to be a function of the UO₂ content. On the basis of these observations, it appears that the UO₂ content of the solid solution fuels is a dominant factor in their high temperature thermal conductivity behavior.

Fig. 7

A comparison of thermal conductivity data reported previously⁽⁵⁾ (Figure 7) with the present data for similar compositions of these homogeneous ZrO₂-base fuels shows good agreement between the low temperature and high temperature test methods. The thermal conductivity up to about 800°C was determined by Materiadyne Corporation using a steady-state comparative method, while the thermal conductivity between 900 to 2200°C was determined at CEND using a high thermal gradient radial heat flow method.

Also in Figure 7 is a comparison of UO₂ thermal conductivity determined by CEND⁽⁸⁾ and GE-NMPO⁽⁷⁾. An exceptionally good correlation is evident. The CEND values were determined from measurements obtained using the same high gradient, radial heat flow technique and equipment described above, while GE-NMPO used a low gradient, essentially isothermal radial heat flow technique for their measurements.

This close correlation of data obtained by two grossly different techniques provides a high degree of confidence in the suitability of the technique for making such measurements.

These new thermal conductivity data are being utilized to provide a new series of surface temperature-fuel centerline temperature relationships which are vital to the controlled movement of the irradiation test capsules.

b. Effective Thermal Conductivity

The calculated pellet inside surface temperature and the exterior cladding temperature were used in making the calculations for effective thermal conductivity of the test elements. The results of these

calculations as a function of pellet inside surface temperature are presented in Figure 8. Density corrections were not made. The shape of the k_{eff} curves are similar to the thermal conductivity curves shown in Figures 6 and 7. The expected lower values for k_{eff} are a result of the interface resistance between the fuel pellets and the cladding. The increase in k_{eff} at elevated temperatures reflects the influence of higher conductivities noted for these fuels at higher temperatures.

These values for k_{eff} are slightly higher than those reported for 4 m/o and 12 m/o UO_2 fuels in CEND-158⁽⁵⁾. The slight difference can be attributed to the following two factors:

(1) The present values of the k_{eff} were determined from the pellet inside surface temperature and clad temperature and were plotted against pellet inside surface temperature whereas, the values of k_{eff} in CEND-158⁽⁵⁾ were determined from heater surface temperature and clad temperature and plotted against heater rod temperature. This effect produces a lower ΔT for any given heat flux, thus increasing the k_{eff} .

(2) These test elements had cold diametral clearances between heater to pellet and pellet to clad of less than 0.003 cm (0.001 inch). Earlier test elements had diametral clearances of about 0.015 cm (0.006 inch) between heater rod and pellets and about 0.010 cm (0.004 inch) between pellets and clad. The higher interfacial conductivity in the present series of tests is reflected in a greater k_{eff} .

**- EFFECTIVE THERMAL CONDUCTIVITY -
COMPARISON OF Z-10 COMPOSITION TO Z-22 COMPOSITION**

Fig. 8

2. Microstructural Changes

Upon completion of thermal cycle testing, the fuel elements were impregnated with a commercial sealant which locked all the fragments in place and permitted sectioning in accordance with details shown in Figure 4. Polishing and etching procedures were similar to those discussed in another section of this report.

Descriptions of microstructures developed in these thermal test elements are discussed below:

a. Element 1: Z-10 Composition

The appearance of the samples from element 1 indicated little microstructural change in the form of grain growth, in densification of the fuel or pore distribution. Since the maximum fuel temperature was about the same as the sintering temperature of the fuel, this test demonstrated that high thermal gradients at maximum fuel temperatures in the range of 1800 to 1850°C for short periods of time did not alter the as-sintered microstructure. Figure 9 shows the appearance of a typical microstructure from element 1 of the Z-10 composition. This photomicrograph shows evidence of intimate contact between fuel and heater rod at test temperatures. A small amount of deterioration of heater rod as well as migration of fine metallic particles into the fuel can also be seen. The uniform grain size and pore distribution of this region very closely resembled the as-sintered Z-10 fuel microstructure (Figure 9).

b. Element 2: Z-22 Composition

Maximum fuel temperatures achieved in this element were about 2300°C for both test cycles. Microscopic examinations showed

2871

AS SINTERED

100X

2868

ELEMENT #1 SECTION 8
POST TEST

100X

TYPICAL AS SINTERED AND POST TEST Z-10 COMPOSITION MICROSTRUCTURES FROM
THERMAL PERFORMANCE SPECIMENS

a very narrow band adjacent to the heater rod where a limited amount of grain growth occurred (Figure 10).

Grain size measurements and the technique for determining onset of grain growth described for UO_2 microstructures in CEND-2863-217⁽⁸⁾ were used in the analysis of these microstructures.

The change in grain size is abrupt, indicated by a sharp delineation between the as-sintered and growth region. Of particular interest is the high density and pore redistribution noted in the growth region when compared to the microstructure in the colder regions of the pellet which resembles that of the as-sintered microstructure (Figure 10). From the appearance of these microstructures (Figures 9 and 10) it is evident that grain growth (and associated redistribution of voids and densification) under short time, high thermal gradient conditions does not occur unless fuel temperatures greater than 2000°C are attained. When this temperature is exceeded, a growth region is formed which is essentially free of voids.

The microstructure shown in Figure 11 was produced at maximum fuel temperatures within 50 to 100°C of the melting point (2635°C)⁽⁵⁾ of the Z-10 composition. An analysis of the end-of-life thermal history and the post-test microstructure, as described earlier⁽⁸⁾, established the radial temperature isotherm of about 1900°C as the onset of grain growth. The large blocky-columnar grains produced under these conditions are almost completely free of voids. These changes in microstructure further substantiates the earlier observation concerning the requirement for temperatures in excess of 2000°C to initiate discontinuous grain growth in these fuels under thermal gradient conditions.

2869

AS SINTERED

100X

2870

ELEMENT #2 SECTION 7
POST TEST

100X

TYPICAL AS SINTERED AND POST TEST Z-22 COMPOSITION MICROSTRUCTURES FROM
THERMAL PERFORMANCE SPECIMENS

2872

Z-10 COMPOSITION

100 X

*MICROSTRUCTURES FROM Z-10 COMPOSITION
THERMAL PERFORMANCE SPECIMEN SHOWING
GRAIN GROWTH*

Fig. 11

Long term isothermal heat treatment (reported in detail in Section V) at 1400°C has produced massive grain growth in the ZrO_2 -base fuels. Figures 22 and 23 show the microstructural changes in Z-10 and Z-22 specimens after 326 days at temperatures between 1400 and 1475°C . It would appear that the temperature required to produce grain growth under isothermal conditions is several hundred degrees centigrade below the estimated temperature for grain growth under high thermal gradient conditions. However, the presence of the low melting point glassy phase in the isothermal samples enhanced grain growth by means of liquid phase sintering, thus complicating comparisons.

Grain growth under high thermal gradient conditions produced large blocky grains (Figure 11) which were highly densified and almost completely free of porosity, whereas isothermal conditions produce coarsening of the grains and increased pore size with very little apparent void redistribution, consistent with the above observation concerning enhanced liquid phase sintering. On this basis, it is concluded that the temperatures noted above for the onset of discontinuous, large blocky microstructure constituents can be used as a basis for post-irradiation evaluations.

IV. CORROSION TESTS

A. Introduction

The corrosion testing of unclad fuel pellets of ZrO_2 -base and CeO_2 -base fuel compositions, shown in Tables I and II, is continuing. An evaluation of corrosion behavior is necessary in the event of a cladding failure and subsequent exposure of the fuel to the coolant environment. A description of sample preparation methods was given in CEND-158⁽⁵⁾; results of earlier corrosion tests were reported in CEND-158⁽⁵⁾ and CEND-190⁽⁹⁾, the latter following exposures of 300 days in static degassed water at 360°C and 420 days in flowing steam at 650°C . Weight gain measurements indicated corrosion penetrations of less than one mil per year. Visual and metallographic examinations indicated that none of the pellets had developed severe cracks. In addition, heavy surface corrosion products were not evident.

Corrosion testing has been extended to 480 days in 360°C water and to 700 days in 650°C steam. Results of evaluations are reported below.

B. Results and Discussion

The long term stability of both the ZrO_2 -base and CeO_2 -base fuel pellets in degassed water at 360°C and steam at 650°C is demonstrated in Figures 12 and 13. Corrosion data for all fuel compositions indicate decreasing rates with increasing time. Continued exposure to these test environments did not alter the corrosion behavior or the physical appearance of these samples. The previously reported⁽⁹⁾ corrosion penetration of less than 1 mil per year (based on weight change) was maintained even after exposures of 480 days in

WATER CORROSION DATA AT 360°C

Fig. 12

Fig 13

TABLE I

Composition of Steam Corrosion Test Samples

<u>Pellet Type</u>	<u>UO₂</u>	<u>Mole Percent CaO Stabilized ZrO₂*</u>	<u>CeO₂</u>
1	100	--	--
2	--	--	100
3	--	100	--
4	4	96	--
5	12	88	--
6	15	85	--
7	7	--	93
8	21	--	79

* Containing 20 m/o CaO

TABLE II

Composition of Water Corrosion Test Samples

<u>Pellet Type</u>	<u>UO₂</u>	<u>Mole Percent CaO Stabilized ZrO₂*</u>	<u>CeO₂</u>
1	--	--	100
2	--	100	--
3	7	--	93 (single sinter)
4	17.5	--	82.5
5	7	--	93 (double sinter)
6	4	96	--
7	12	88	--

* Containing 20 m/o CaO

360°C degassed water and 700 days in 650°C steam. These results are especially significant in the case of pellets exposed to flowing steam where the combination of elevated temperature and a dynamic environment constituted a more severe test condition. The corrosion behavior of these solid solution oxide fuels compares favorably with reported⁽¹⁰⁾ data for sintered UO₂ pellets exposed to high temperature water and steam.

Detailed visual examination of the test samples indicated that the ZrO₂-base pellets had retained their previously formed light corrosion film. Shallow, randomly distributed pits were observed at magnifications of 10X and higher, with the pellets of the higher UO₂ composition exhibiting the greater population of pits. There was no visual evidence of physical damage in the form of large cracks, chipped edges or swelling of the pellets. The CeO₂-base pellets were coated with an adherent gray deposit accompanied with shallow attack at the grain boundaries. Surface pitting, up to about 2 mils in depth, was observed to increase with increasing UO₂ content in a manner similar to that noted for the ZrO₂-base pellets. Fine cracks were observed in the pellets containing the higher UO₂ content.

The effect of extended exposures to 360°C water is shown in the microstructure of Figures 14 through 17 for both the as-polished and etched conditions. The appearance of these samples is not consistent with that observed after 300 days in water and 420 days in steam. The present condition of these samples may be a result of a recent autoclave shutdown caused by a ruptured safety disc. The loss of water as well as the associated thermal shock may have been instrumental in inducing the effects shown in Figures 14 through 17. This observation is particularly significant in light of the excellent

2875

UNETCHED

4 M/O UO_2

50X

2876

UNETCHED

12 M/O UO_2

50X

STRUCTURES OF THE ZrO_2 -BASE PELLETS FOLLOWING 480 DAYS EXPOSURE TO 360°C WATER

2831

ETCHED

50X

4 M/O UO_2

2832

ETCHED

50X

12 M/O UO_2

STRUCTURES OF THE ZrO_2 -BASE PELLETS FOLLOWING 480 DAYS EXPOSURE TO 360°C WATER

2873

UNETCHED

7 M/O UO_2

50X

2874

UNETCHED

17.5 M/O UO_2

50X

STRUCTURES OF CeO_2 -BASE PELLETS FOLLOWING 480 DAYS EXPOSURE TO 360° C WATER

2833

ETCHED

7 M/O UO₂

50 X

2834

ETCHED

17.5 M/O UO₂

50 X

STRUCTURES OF CeO₂-BASE PELLETS FOLLOWING 480 DAYS EXPOSURE TO 360° C WATER

appearance of the pellets tested in steam as shown in Figures 18 and 19.

Detailed metallographic analyses of the samples tested in water indicate that compositions in both oxide systems showed increased susceptibility to cracking with increasing UO_2 content. The interior of the ZrO_2 -base pellets had undergone localized intergranular separation resulting in the removal of single grains during the mechanical preparation of the specimens. The intergranular nature of the attack is shown in the etched microstructures of Figure 15. The locations of removed grains indicated that grain boundary separation is probably not related to the intergranular cracks.

The CeO_2 -base compositions exhibited extremes in physical behavior. The 7 m/o UO_2 pellet was free from cracks and intergranular attack whereas the 21 m/o UO_2 pellet had developed extensive networks of fine intergranular and transgranular cracks. (See Figures 16 and 17.)

Similar microscopic examination of the ZrO_2 -base pellets from the 650°C steam corrosion test showed several fine transgranular cracks which were oriented chiefly in the radial direction. There was no evidence of intergranular attack nor grain boundary separation. The CeO_2 -base pellets containing 7 m/o and 21 m/o UO_2 , were free from cracks and intergranular attack. In general, there were no significant changes in the structural quality of all pellets as a result of additional exposures to 650°C steam.

A comparison of microstructures of identical composition pellets from both tests indicated that only the 360°C water tested pellets had cracked to a significant extent. This may be attributed to the recent shutdown of the autoclave. The suspected recent origin of the cracks, especially in the CeO_2 -base pellet, is supported by the complete absence of swelling and crumbling.

2877

ETCHED

4 M/O UO_2

50X

2878

ETCHED

12 M/O UO_2

50X

STRUCTURES OF THE ZrO_2 -BASE PELLETS FOLLOWING 700 DAYS EXPOSURE TO 650°C STEAM

2879

ETCHED

7 M/O UO_2

50X

2880

ETCHED

17.5 M/O UO_2

50X

STRUCTURES OF THE CeO_2 -BASE PELLETS FOLLOWING 700 DAYS EXPOSURE TO 650° C STEAM

Testing of pellets in 360°C water and in 650°C flowing steam will be continued with periodic examinations each 90 days. Specific emphasis will be placed on the behavior in the water tests of pellets which appear to be in an initial stage of deterioration. Test data will be utilized in comparing corrosion behavior of irradiated pellets of the same compositions which will be conducted during the final phase of the irradiation program.

The long-time corrosion tests have demonstrated the excellent resistance of both the ZrO₂-base and CeO₂-base fuels to high temperature water or steam. Cracking of pellets of high UO₂ content appear to be of recent origin and may be due to thermal shock during an emergency shutdown of the autoclave. None of the pellets exposed to 650°C steam exhibited similar cracks. On the basis of these results, good fuel integrity should be maintained in either water or steam environments for periods of time approaching one core lifetime in the event of a cladding defect.

V. THERMAL STABILITY

A. Introduction

The second type of out-of-pile thermal tests has involved long-time heat treatments of pellets of the four fuel compositions to distinguish the thermal effects from the irradiation effects as an aid in the interpretation of post-irradiation data. A temperature of 1400 to 1475°C was selected on the basis of previous shorter-time (28 days) phase stability investigations⁽⁵⁾ which indicated that single phase structures, formed after sintering at 1800°C, transformed to two phase structures at 1300 and 1500°C for $\text{UO}_2\text{-CaO-ZrO}_2$ compositions containing greater than 12 m/o UO_2 . Thus, the selection of 1400 to 1475°C as the temperature for these long-time thermal stability investigations for the 4 and 10.2 m/o UO_2 compositions provided the maximum potential for transformation, if transformations were to occur.

Since the irradiation test samples were subjected to a series of thermal cycles during normal test reactor operation and shutdown, the influence of cycling on phase stability was evaluated by a series of thermal cycles at temperatures below 1400 to 1475°C. Although little influence of cycling was anticipated if the structures remained single phase, the influence of cycling could not be predicted if multi-phase structures were developed.

A comparison of the irradiated and out-of-pile heated structures of the same fuel compositions would assist in distinguishing irradiation induced changes (segregation of fission products, precipitation of fission gases into voids) from pure thermal effects (grain size and transformation phases, if any).

B. Experimental

Unclad pellets of the two ZrO_2 -base compositions were heat treated in a vertical furnace at temperatures ranging from 1400 to 1475°C. The pellets were supported on a stabilized ZrO_2 rod which extended from the bottom of the furnace into the hot zone. A silicon carbide resistance element externally heated the stabilized ZrO_2 furnace tube containing the support rod and pellets. A Pt-Pt·Rh thermocouple extended through an axial hole within the support rod for measurement of temperatures of the pellets. Variations in pellet temperatures were obtained by raising or lowering the support rod to various levels within the furnace as well as by controlling the power input. Argon (99.99% purity) was flowed at a slight positive pressure through the furnace tube during the heat treatments. During furnace shutdowns, the pellets were removed for examinations by lowering the support rod completely out of the furnace.

Heat treatments of the CeO_2 -base fuel pellets were conducted in a horizontal Zircon tube furnace which was maintained at temperatures of 1380 to 1415°C. The lower heat treat temperature was selected for the following reasons:

1. In-pile temperatures of the CeO_2 -base fuels are lower than those for the ZrO_2 -base fuels.
2. Significant losses of fuel by volatilization would occur during long-time heat treatments above about 1450°C.

Pure argon was flowed through the furnace tube at slightly above atmospheric pressure. A Pt-Pt·Rh thermocouple was positioned within the region of the pellets for periodic temperature measurements. The furnaces

were cooled at a rate of about 100°C per hour to prevent thermal shock cracking of the pellets.

A series of thermal cycles to room temperature was conducted on the pellets within both furnaces to obtain interim information on the physical behavior and dimensional stability of the pellets. These scheduled furnace shutdowns also allowed inspections and replacement of heaters and furnace tubes. The ZrO₂-base pellets have completed 326 days exposure at the elevated temperatures while the CeO₂-base pellets have attained 256 days (Figures 20 and 21). At the end of these test periods, all pellets were examined visually, dimensioned and weighed. One pellet of each of the four fuel compositions was evaluated for microstructure and crystal structure. The remaining pellets were retained for continued high temperature heat treatments.

Sample preparation for microstructural examination were conducted by polishing and etching techniques described in CEND-190⁽⁹⁾. Photographic traverses at 100X of the ZrO₂-base pellets were made on unetched samples since the grain boundaries were clearly delineated by the presence of an impurity glassy phase. This impurity phase was not detected in the pellets before test. The cause for the development of this impurity phase will be discussed later in this section. Representative areas were photographed. The CeO₂-base samples were essentially free from the glassy material and the grain structures were delineated by etching for photographic traverses at 100X and for the high magnification views at 500X.

Samples for X-ray diffraction analyses were removed from both the surface and the interior of the ZrO₂-base pellets and crushed to a fine particle size in helium to prevent oxidation. The X-ray cameras were filled

THERMAL STABILITY TEST — Z-10 and Z-22 COMPOSITIONS

Fig. 21

THERMAL STABILITY TEST — C10 and C25 COMPOSITIONS

with helium during film exposures. In view of possible compositional changes at the pellet surfaces, the structure of the interior of the pellets was used as the basis for evaluations.

C. Results and Discussion

Descriptions of the appearance of heat treated pellets are included in Table III. Apparent changes in composition had occurred in the ZrO_2 -base pellets in a shallow surface layer evidenced by formation of a tan colored translucent surface region. Losses of UO_2 by volatilization and redeposition were evident by the formation of an orange to brown-colored reaction zone in the furnace tube at the location of the pellets. Similarly, volatile losses of CeO_2 -base pellets were indicated by black crystalline deposits on the inside surfaces of the furnace tube. These deposits were identified to be U_3O_8 by X-ray diffraction. The fuel pellets had lost U_3O_8 during heat treatment by one of two mechanisms:

1. Disproportionation of the high UO_2 composition with the formation of U_3O_8 and a CeO_2 -rich phase with the resulting volatilization of U_3O_8 .
2. Preferential loss of UO_2 in the form of U_3O_8 from the surfaces of the pellets.

It will be shown later that the structural changes noted above were limited to a narrow surface layer and that the interior of the pellets was not influenced by surface volatilization.

The microstructures of the ZrO_2 -base compositions are shown in diametral traverses of Figures 22 and 23. ZrO_2 -base pellets containing 4 m/o UO_2 had developed a large grain size (220 microns) with an essentially continuous

MICROSTRUCTURE OF HEAT TREATED Z-10 FUEL PELLET

MICROSTRUCTURE OF HEAT
TREATED Z-22 FUEL PELLET

Fig. 23

TABLE III

Appearance of Heat Treated Pellets

Fuel Composition

External Appearance

Z10

All pellets exhibited a tan-colored surface layer which was thermally etched at the grain boundaries. Interior of pellets was dark brown to gray in color. There was no evidence of cracks, chipped edges or other abnormalities.

Z22

External appearance resembled that of Z10 pellets except for more severe thermal etching at the grain boundaries.

C10

Uniformly black with no evidence of cracks, chipped edges or other abnormalities. Grain boundaries lightly thermally etched with slight pitting.

C25

External appearance similar to that of C10 pellets except for severe erosion and pitting at the grain boundaries.

glassy phase* at the grain boundary (Figure 22). The porous surface layer, extending to 0.127 cm (0.050 inch) in depth, showed a high concentration of the glassy material. The large grain size may, in part, be attributed to the presence of a liquid phase at the grain boundaries during the heat treatments. The fuel structures for Z-10 samples had remained thermally stable with no evidence of minor phases within the grains. A metallic-like constituent reported for the irradiated fuel of the same composition was not observed in the out-of-pile heat treated pellets. The structure of the 10.2 m/o UO_2 composition exhibited similar trends of grain growth (180 microns) with the formation of a grain boundary glassy phase. Trace amounts of an unidentified fine precipitate was observed within the grains (Figure 24). None of the pellets showed the "annular rings" reported for the irradiated fuel pellets of these compositions.

The CeO_2 -base pellets also exhibited single phase structures following the long-time heat treatments. Typical microstructures are shown in the diametral traverses of Figures 25 and 26. The use of high purity BeO as the pellet support material was effective in preventing the introduction of low melting point impurity materials as in the case of the ZrO_2 -base pellets. The phenomenon of grain growth during extended heat treatments is shown in the microstructures of the CeO_2 -base compositions (Figures 27 and 28). Grain growth is characterized by the disappearance of some of the grains by migration

*The glassy material was not present in the pellets prior to test and is believed to consist of impurities (SiO_2 , Al_2O_3 , TiO_2 , MgO and Fe_2O_3) from the stabilized ZrO_2 furnace tube and support rod. The higher concentration of impurity phase at the surface of pellets of both zirconia-base compositions support the proposed method of external pickup from the furnace tube and support rod. Evidence of this phase was not obtained on X-ray diffraction photograms.

*FINE PRECIPITATE WITHIN GRAINS OF
HEAT TREATED Z22 STRUCTURE*

MICROSTRUCTURE OF HEAT
TREATED C-10 FUEL PELLETS

Fig 25

MICROSTRUCTURE OF HEAT
TREATED C-25 FUEL PELLET

Fig 26

ETCHED 500 X
STRUCTURE OF C-10 (7M/O UO_2) COMPOSITION FOLLOWING 256 DAYS
OF HEAT TREATMENT

Fig. 27

ETCHED 500 X
STRUCTURE OF C-25 (17.5M/O UO_2) COMPOSITION FOLLOWING 256 DAYS
OF HEAT TREATMENT

Fig. 28

of grain boundaries toward their centers of curvature. The parallel traces in the photomicrographs mark the progressive positions of the migrating grain boundaries at each furnace cooldown. A similar microstructure attributable to this phenomenon was observed in the irradiated C-25 fuel.

Results of X-ray diffraction analyses of the heat treated samples are presented in Table IV together with lattice constants.

TABLE IV
X-Ray Diffraction Data of Heat Treated Samples

<u>Composition</u>	<u>Sample Location</u>	<u>Structure</u>	<u>Lattice Constant - Å</u>	
			<u>Before Heat Treatment</u>	<u>After Heat Treatment</u>
Z10	Surface	Single phase FCC	---	5.134
	Interior	Single phase FCC	5.149	5.151
Z22	Surface	Single phase FCC	---	5.133
	Interior	Single phase FCC	5.173	5.168
C10	Surface	Single phase FCC	---	5.375
	Interior	Single phase FCC	5.415	5.413
C25	Surface	Single phase FCC	---	5.408
	Interior	Single phase FCC	5.421	5.419

All X-ray diffraction patterns were sharp, indicating equilibrium structures. As anticipated, the minute precipitate observed in the Z-22 structure was not detected in the X-ray diffraction photograms.

Lattice constant-composition relationships developed earlier in the program along with current data are shown in Figure 29*. These data will be useful in the crystallographic work, currently in progress, on the irradiated ZrO_2 -base fuels. The information of Table IV and Figure 29 were employed to establish the extent of UO_2 losses from the surfaces of the heat treated pellets. It is seen from Figure 29 that the lattice constant of 5.133 to 5.134 Å for the pellet surfaces corresponds to a UO_2 content of 2.4 m/o, amounting to a loss of 1.6 m/o UO_2 for the Z-10 pellet and 7.8 m/o UO_2 in the case of the Z-22 pellet. Both X-ray diffraction and microscopy examinations indicate that the two ZrO_2 -base compositions exhibited excellent thermal stability following the extended heat treatment. The fine precipitate in the 10.2 m/o UO_2 composition may be an impurity phase rather than a decomposition product, however, this hypothesis has not been confirmed.

The single phase, face centered cubic structures of the two CeO_2 -base compositions remained unchanged as a result of the heat treatment. Additional phases were not detected in the microstructure. It is concluded that the loss of UO_2 by volatilization had occurred preferentially from the surface of the pellets rather than by phase transformation. This is evident by the highly porous surfaces in the structures shown in Figures 25 and 26.

D. Conclusions

1. The single phase, face centered cubic structures in the ZrO_2 -base fuel compositions remained thermally stable following 326 days at 1400 to 1475°C in argon. A faint trace of fine precipitate within the grains of the 10.2

*Included also are data obtained for the Phillips Petroleum Company as part of the Atomic Energy Commission Power Burst Program.

LATTICE CONSTANT Zr O₂-BASE FUEL COMPOSITIONS

Fig.29

m/o UO_2 composition was not identified and could only be observed after careful metallographic preparation. It is probable that this is a minor impurity phase, originally present in starting materials which agglomerate after long-time heat treatment. Loss of UO_2 by volatilization was limited to a shallow surface layer in pellets of both composition. Low melting point impurities from the furnace tube and support rod resulted in a grain boundary glassy phase.

2. The CeO_2 -base fuel compositions exhibited single phase, face centered cubic structures after 256 days at 1380 to 1450°C in argon. Losses of UO_2 by volatilization had occurred from a shallow surface layer of pellets of the two compositions.

3. Grain size measurements show that the lower UO_2 compositions in both fuel systems showed a greater tendency for grain growth. It is possible that the presence of the glassy phase in the ZrO_2 -base pellets may have caused an exaggerated grain size.

VI. IRRADIATION TESTING

A. Introduction

This Section of the report summarizes progress on the irradiation testing of the six capsules in the ETR and MTR. Two capsules, 43-1 and 43-4, were discharged during April 1964 after attaining fission burnups of 40% of the U-235 atoms. The irradiation testing of the remaining four capsules is continuing in the ETR to burnup levels of 55 and 70% of the U-235 atoms. Results of hot cell examinations and preliminary data on burnup analyses are included. Although no definitive conclusions could be made on fuel performance at the present time, all pertinent data are presented. An interim report will be prepared early in 1965 to provide complete post-irradiation analyses of fuels from these two capsules.

B. Irradiation

Irradiation testing of the six capsules, described previously⁽⁹⁾, was continued. Target irradiation conditions for each capsule are shown in Table V. In an attempt to maintain a relatively constant centerline temperature as fuel burnup progressed, scheduled capsule moves have been made to permit all capsules to operate at their maximum heat generation rates consistent with the requirement that the melting point is not exceeded. The general scheme which was adopted was to move a capsule to a higher flux position when the calculated centerline temperature of the fuel was 540°C below the target temperature established for each fuel type. It was necessary to modify this idealistic approach on the basis of available positions in the test reactors. In some instances, movement of capsules to reportedly higher flux positions had

TABLE V

Target Irradiation Conditions

Capsule Designation	Fuel Designation ^(a)	Target Specimen Temperatures, °F		Target Specimen Burnups, a/o of ²³⁵ U by Fission	Fissions/cm ³ x 10 ²⁰
		Clad Outer Surface	Specimen Center		
43-1	Z-10	600	4000	40	4.4
	C-10	600	3400		6.1
43-2	Z-10	600	4000	55	6.1
	C-10	600	3400		8.3
43-3	Z-10	600	4000	70	7.7
	C-10	600	3400		10.6
43-4	Z-22	600	4000	40	10.5
	C-25	600	3400		15.3
43-5	Z-22	600	4000	70	18.4
	C-25	600	3400		27.0
43-6	Z-22	600	4000	55	14.4
	C-25	600	3400		21.0

(a) Z-10 and C-10 fuels contain 10 wt% UO₂; Z-22 and C-25 fuels contain 22 wt% and 25 wt% UO₂, respectively.

little influence on their thermal performance. When such conditions were observed, additional moves were made. In all instances, attempts were made to achieve maximum heat generation rates consistent with the above ground rules. In some cases, it was necessary to retain existing capsule locations, even though heat generation rates were below target values, on the basis that movement to the next available higher flux position would have resulted in temperatures exceeding those of the melting point of the fuel. In two instances, excessive temperatures were reached while the reactor was at reduced power. The latter instances occurred during reactor start-up for ETR cycles 58 and 60 for capsule 43-6. It was necessary, in both cases, to shut the reactor down and move the capsules to a lower flux.

Sixteen capsule relocations were performed during this reporting period. Tables VI through XI indicate all capsule relocations together with average fuel centerline temperatures for each cycle in each position. These data are plotted in Figures 30 through 33. As shown in Figure 33, target maximum centerline temperatures were again achieved for 43-4 during the final two cycles as a result of a programmed relocation. Thus, post-irradiation evaluations are now based on fuel that has been subjected to target maximum thermal conditions and at target burnups for this capsule. In the case of capsule 43-1, a capsule relocation was not possible to provide the maximum target thermal conditions at the end of irradiation test life. In reviewing future anticipated moves, it was concluded that such moves would not be possible for a number of cycles. On this basis, this capsule was discharged at the design burnup having operated at fuel centerline temperatures in the lower range of the target limits

Fig. 30

AVERAGE FUEL CENTERLINE TEMPERATURE vs. ETR CYCLE NUMBER

AVERAGE FUEL CENTERLINE TEMPERATURE vs. BURNUP

AVERAGE FUEL CENTERLINE TEMPERATURE vs. MTR CYCLE NUMBER

AVERAGE FUEL CENTERLINE TEMPERATURE vs. BURNUP

TABLE VI

Irradiation Data Summary for Capsule 43-1

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
57	P-13-NE	Z-10-A	450	2700	2.8	23.3
		C-10-A	490	2350	2.9	24.7
		Z-10-B	480	2950	3.0	25.7
		Z-10-C	490	3050	3.1	26.1
		C-10-B	450	2150	2.7	21.9
58	P-12-SE	Z-10-A	490	3000	2.7	26.0
		C-10-A	540	2600	2.9	27.6
		Z-10-B	520	3300	2.9	28.6
		Z-10-C	540	3400	3.0	29.1
		C-10-B	510	2400	2.7	24.6
59	P-12-SE	Z-10-A	470	2850	3.2	29.2
		C-10-A	510	2450	3.5	31.1
		Z-10-B	530	3300	3.8	32.4
		Z-10-C	510	3150	3.6	32.7
		C-10-B	470	2200	3.0	27.6
60	P-12-SE	Z-10-A	470	2850	3.7	32.9
		C-10-A	520	2500	4.0	35.1
		Z-10-B	520	3300	4.3	36.7
		Z-10-C	510	3200	4.1	36.8
		C-10-B	480	2250	3.6	31.2

TABLE VI (Continued)

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
61	P-12-SE	Z-10-A	450	2750	3.4	36.3
		C-10-A	500	2400	3.8	38.9
		Z-10-B	510	3150	4.0	40.7
		Z-10-C	500	3050	3.9	40.7
		C-10-B	460	2150	3.4	34.6
62	P-12-NE	Z-10-A	480	2900	3.7	40.0
		C-10-A	410	1850	2.9	41.8
		Z-10-B	510	3200	4.0	44.7
		Z-10-C	490	3050	3.9	44.6
		C-10-B	450	2100	3.2	37.8

TABLE VII

Irradiation Data Summary for Capsule 43-2

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
9-1A	P-6-SE	Z-10-D	580	3750	3.8	14.2
		Z-10-E	600	3900	4.0	14.7
		Z-10-F	630	4100	4.3	15.5
		C-10-C	630	3150	4.0	14.8
		C-10-D	560	2750	3.5	12.4
	P-6-SE	Z-10-D	510	3150	2.8	17.0
		Z-10-E	530	3300	3.0	17.7
		Z-10-F	560	3550	3.2	18.7
		C-10-C	560	2750	3.0	17.8
		C-10-D	500	2400	2.6	15.0
	P-6-SE	Z-10-D	520	3250	3.7	20.7
		Z-10-E	530	3350	3.8	21.5
		Z-10-F	540	3450	3.9	22.6
		C-10-C	550	2650	3.8	21.6
		C-10-D	490	2300	3.3	18.3
	P-6-SE	Z-10-D	560	3600	4.7	25.4
		Z-10-E	590	3750	5.0	26.5
		Z-10-F	600	3850	5.1	27.7
		C-10-C	610	3050	4.9	26.5
		C-10-D	530	2600	4.2	22.5

TABLE VII (Continued)

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
57	P-6-SE	Z-10-D	580	3750	3.8	14.2
		Z-10-E	600	3900	4.0	14.7
		Z-10-F	630	4100	4.3	15.5
		C-10-C	630	3150	4.0	14.8
		C-10-D	560	2750	3.5	12.4
58	P-6-SE	Z-10-D	510	3150	2.8	17.0
		Z-10-E	530	3300	3.0	17.7
		Z-10-F	560	3550	3.2	18.7
		C-10-C	560	2750	3.0	17.8
		C-10-D	500	2400	2.6	15.0
59	P-6-SE	Z-10-D	520	3250	3.7	20.7
		Z-10-E	530	3350	3.8	21.5
		Z-10-F	540	3450	3.9	22.6
		C-10-C	550	2650	3.8	21.6
		C-10-D	490	2300	3.3	18.3
60	P-6-SE	Z-10-D	560	3600	4.7	25.4
		Z-10-E	590	3750	5.0	26.5
		Z-10-F	600	3850	5.1	27.7
		C-10-C	610	3050	4.9	26.5
		C-10-D	530	2600	4.2	22.5
61	P-6-SE	Z-10-D	520	3250	4.1	29.5
		Z-10-E	540	3400	4.3	30.8
		Z-10-F	510	3150	3.9	31.6
		C-10-C	520	2500	4.0	30.5
		C-10-D	490	2350	3.6	26.1

TABLE VII (Continued)

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
62	P-6-SE	Z-10-D	510	3200	4.0	33.5
		Z-10-E	520	3250	4.1	34.9
		Z-10-F	490	3050	3.9	35.5
		C-10-C	530	2600	4.1	34.6
		C-10-D	480	2250	3.5	29.6
63	P-6-SE	Z-10-D	510	3200	4.7	38.2
		Z-10-E	520	3250	4.7	39.6
		Z-10-F	520	3250	4.7	40.2
		C-10-C	520	2500	4.5	39.1
		C-10-D	460	2150	3.9	33.5
64	P-5-SW	Z-10-D	490	3000	2.9	41.1
		Z-10-E	510	3150	3.1	42.7
		Z-10-F	510	3200	3.2	43.3
		C-10-C	510	2500	3.0	42.1
		C-10-D	460	2150	2.6	36.1

TABLE VIII

Irradiation Data Summary for Capsule 43-3

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
6-IA	G-17-NE	Z-10-G	460	3050	3.2	10.0
		Z-10-H	470	3100	3.2	10.0
		Z-10-J	510	3450	3.6	11.5
		C-10-E	500	2600	3.4	10.8
		C-10-F	470	2400	3.1	9.8
	F-16-SE	Z-10-G	470	3100	2.8	12.8
		Z-10-H	460	3000	2.7	12.7
		Z-10-J	510	3400	3.1	14.6
		C-10-E	400	1900	2.1	12.9
		C-10-F	480	2450	2.8	12.6
	F-16-SE	Z-10-G	520	3550	4.1	16.9
		Z-10-H	540	3700	4.2	16.9
		Z-10-J	580	4050	4.7	19.3
		C-10-E	560	3000	4.3	17.2
		C-10-F	570	3050	4.4	17.0
	F-16-SE	Z-10-G	480	3200	4.2	21.1
		Z-10-H	500	3300	4.4	21.3
		Z-10-J	560	3800	5.0	24.3
		C-10-E	540	2850	4.7	21.9
		C-10-F	570	3000	4.9	21.9

TABLE VIII (Continued)

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
61	F-16-SE	Z-10-G	500	3300	4.2	25.3
		Z-10-H	510	3400	4.4	25.7
		Z-10-J	570	3950	5.0	29.4
		C-10-E	570	3050	4.8	26.7
		C-10-F	580	3100	5.0	26.9
62	F-16-SE	Z-10-G	490	3250	4.2	29.5
		Z-10-H	500	3350	4.3	30.0
		Z-10-J	560	3850	4.9	34.4
		C-10-E	550	2900	4.6	31.4
		C-10-F	560	2950	4.7	31.6
63	F-16-SE	Z-10-G	460	3050	4.5	34.0
		Z-10-H	470	3100	4.6	34.6
		Z-10-J	530	3600	5.4	39.8
		C-10-E	520	2750	5.0	36.4
		C-10-F	530	2750	5.0	36.6
64	G-16-SW	Z-10-G	570	3950	3.9	37.9
		Z-10-H	570	3950	3.9	38.5
		Z-10-J	620	4400	4.4	44.2
		C-10-E	570	3050	3.7	40.1
		C-10-F	550	2900	3.6	40.2

TABLE IX

Irradiation Data Summary for Capsule 43-4

MTR Cycle Number	MTR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
195	A-29-SE	Z-22-M	580	3800	1.8	21.9
		C-25-M	540	2800	1.6	20.1
		Z-22-N	560	3600	1.7	21.4
		Z-22-O	580	3800	1.8	22.4
		C-25-O	570	3000	1.8	21.5
196	A-29-SE	Z-22-M	480	3000	1.4	23.3
		C-25-M	480	2400	1.4	21.5
		Z-22-N	540	3450	1.6	23.0
		Z-22-O	560	2700	1.7	24.1
		C-25-O	560	2950	1.7	23.2
197	A-29-SE	Z-22-M	590	3900	1.9	25.2
		C-25-M	540	2800	1.7	23.2
		Z-22-N	540	3500	1.7	24.7
		Z-22-O	570	3750	1.8	25.9
		C-25-O	570	3000	1.8	25.0
198	A-29-SE	Z-22-M	580	3800	1.8	27.0
		C-25-M	520	2700	1.6	24.8
		Z-22-N	530	3450	1.7	26.4
		Z-22-O	560	3650	1.8	27.7
		C-25-O	560	2950	1.8	26.8

TABLE IX (Continued)

MTR Cycle Number	MTR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
199	A-29-SE	Z-22-M	520	3350	1.4	28.4
		C-25-M	490	2450	1.3	26.1
		Z-22-N	530	3400	1.4	27.8
		Z-22-O	550	3550	1.5	29.2
		C-25-O	540	2800	1.5	28.3
200	A-29-SE	Z-22-M	490	3100	1.7	30.1
		C-25-M	470	2350	1.6	27.7
		Z-22-N	510	3250	1.8	29.6
		Z-22-O	540	3500	1.9	31.1
		C-25-O	540	2800	1.9	30.2
201	A-29-SE	Z-22-M	530	3400	1.6	31.7
		C-25-M	500	2550	1.5	29.2
		Z-22-N	490	3100	1.4	31.0
		Z-22-O	470	2950	1.3	32.4
		C-25-O	450	2250	1.3	31.5
202	A-28-NE	Z-22-M	530	3450	1.5	33.2
		C-25-M	520	2650	1.5	30.7
		Z-22-N	520	3350	1.5	32.5
		Z-22-O	500	3200	1.5	33.9
		C-25-O	480	2400	1.3	32.8
203	A-28-NE	Z-22-M	500	3150	1.6	34.8
		C-25-M	480	2450	1.5	32.2
		Z-22-N	490	3100	1.6	34.1
		Z-22-O	490	3100	1.5	35.4
		C-25-O	470	2350	1.5	34.3

TABLE IX (Continued)

MTR Cycle Number	MTR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
204	A-28-NE	Z-22-M	470	2900	1.3	36.1
		C-25-M	480	2400	1.4	33.6
		Z-22-N	480	3000	1.3	35.4
		Z-22-O	470	2950	1.3	36.7
		C-25-O	450	2250	1.3	35.6
205	A-28-NE	Z-22-M	490	3050	1.5	37.6
		C-25-M	460	2300	1.4	35.0
		Z-22-N	440	2700	1.4	36.8
		Z-22-O	410	2450	1.3	38.0
		C-25-O	390	1850	1.1	36.7
206	A-2-NE	Z-22-M	570	3700	1.5	39.1
		C-25-M	590	3100	1.6	36.6
		Z-22-N	600	4000	1.6	38.4
		Z-22-O	570	3700	1.5	39.5
		C-25-O	540	2800	1.4	38.1
207	A-2-NE	Z-22-M	520	3300	1.3	40.4
		C-25-M	550	2850	1.4	38.0
		Z-22-N	560	3600	1.4	39.8
		Z-22-O	530	3400	1.3	40.8
		C-25-O	510	2600	1.3	39.4

TABLE X

Irradiation Data Summary for Capsule 43-5

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
57	I-2-SW	Z-22-T	520	3350	2.3	8.8
		Z-22-X	520	3350	2.2	8.7
		Z-22-Y	560	3650	2.5	9.6
		C-25-N	580	3050	2.6	9.8
		C-25-S	530	2750	2.2	9.0
58	I-2-SW	Z-22-T	530	3350	1.9	10.7
		Z-22-X	510	3250	1.8	10.6
		Z-22-Y	550	3550	2.0	11.6
		C-25-N	570	3000	2.2	12.0
		C-25-S	520	2650	2.0	11.0
59	I-2-SW	Z-22-T	550	3600	2.7	13.4
		Z-22-X	570	3750	2.7	13.3
		Z-22-Y	570	3700	2.8	14.4
		C-25-N	580	3050	2.9	14.9
		C-25-S	530	2750	2.5	13.5
60	I-2-SW	Z-22-T	540	3450	2.9	16.3
		Z-22-X	540	3450	3.0	16.3
		Z-22-Y	550	3550	3.0	17.4
		C-25-N	570	3000	3.1	18.0
		C-25-S	530	2750	2.9	16.4

TABLE X (Continued)

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
61	I-2-SW	Z-22-T	510	3200	2.7	19.0
		Z-22-X	470	2950	2.4	18.7
		Z-22-Y	490	3050	2.5	19.9
		C-25-N	510	2600	2.6	20.7
		C-25-S	480	2450	2.5	18.9
62	I-3-NW	Z-22-T	590	3850	3.2	22.2
		Z-22-X	620	4100	3.4	22.1
		Z-22-Y	620	4100	3.5	23.4
		C-25-N	630	3400	3.6	24.3
		C-25-S	590	3150	3.3	22.2
63	I-2-SW	Z-22-T	500	3150	3.0	25.2
		Z-22-X	500	3150	3.0	25.1
		Z-22-Y	510	3250	3.0	26.4
		C-25-N	510	2600	3.1	27.4
		C-25-S	480	2400	2.8	25.0
64	I-3-NW	Z-22-T	500	3200	2.0	27.2
		Z-22-X	470	2900	1.8	26.9
		Z-22-Y	520	3300	2.1	28.5
		C-25-N	570	2950	2.4	29.8
		C-25-S	550	2850	2.4	27.4

TABLE XI
Irradiation Data Summary for Capsule 43-6

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
57	Q-12-SW	Z-22-P	490	3050	2.1	16.4
		C-25-P	490	2450	2.1	16.9
		Z-22-R	490	3050	2.0	17.2
		Z-22-Z	480	3000	2.0	16.5
		C-25-R	430	2100	1.8	14.5
58 ^(a)	Q-12-NW	Z-22-P	490	3050	1.8	18.2
		C-25-P	490	2450	1.8	18.7
		Z-22-R	420	3500	1.5	18.7
		Z-22-Z	480	3000	1.7	18.7
		C-25-R	420	2050	1.5	16.0
59	Q-12-NW	Z-22-P	480	3000	2.2	20.4
		C-25-P	480	2400	2.2	20.9
		Z-22-R	450	2750	2.0	20.7
		Z-22-Z	460	2900	2.1	20.3
		C-25-R	410	1950	1.8	17.8
60 ^(b)	P-13-NE	Z-22-P	610	4050	3.4	23.8
		C-25-P	620	3350	3.6	24.5
		Z-22-R	590	3850	3.3	24.0
		Z-22-Z	590	3850	3.3	23.6
		C-25-R	550	2850	3.0	20.8
61	P-13-NE	Z-22-P	620	4100	3.4	27.2
		C-25-P	630	3400	3.5	28.0
		Z-22-R	580	3850	3.2	27.2
		Z-22-Z	580	3850	3.2	27.2
		C-25-R	550	2900	2.9	23.8

TABLE XI (Continued)

ETR Cycle Number	ETR Facility	Specimen Number	Estimated Average Temperatures, °F		Estimated Burnup, a/o of ²³⁵ U by fission	
			Clad Outer Surface	Specimen Center	Accrued	Accumulated
62	P-13-NE	Z-22-P	610	4050	3.4	30.6
		C-25-P	620	3300	3.5	31.5
		Z-22-R	600	3900	3.2	30.4
		Z-22-Z	560	3600	3.0	29.8
		C-25-R	510	2600	2.7	26.5
63	P-13-NE	Z-22-P	620	4100	3.9	34.5
		C-25-P	620	3350	3.9	35.4
		Z-22-R	610	4050	3.9	34.3
		Z-22-Z	570	3750	3.6	33.4
		C-25-R	520	2650	3.2	29.7
64	P-13-NE	Z-22-P	540	3500	2.3	36.8
		C-25-P	540	2800	2.3	37.7
		Z-22-R	540	3450	2.2	36.5
		Z-22-Z	500	3150	2.0	35.4
		C-25-R	450	2250	1.8	31.5

- (a) Capsule was initially inserted into P-12-NE for cycle 58. During startup for cycle 58, a thermocouple temperature of 665°F was attained on specimen Z-22-Z at 156 mw. Therefore, reactor was shut down, and the capsule moved to Q-12-NW for the rest of the cycle.
- (b) Capsule was initially inserted into P-13-SW for cycle 60. During startup for cycle 60, a thermocouple temperature of 650°F was attained on specimen C-25-P at 122 mw. Reactor was shut down, and the capsule moved to P-13-NE for the rest of the cycle.

set up for the program. However, maximum fuel centerline temperatures of 2349°C (4260°F) and 2171°C (3940°F) were noted for sample Z-10-B in this capsule at about one-half and three-quarters of irradiation life, as shown in Table XII. Table XII further identifies the maximum temperatures achieved for test samples during each burnup quadrant. These data are vital to the interpretation of fission gas release and swelling.

Twelve out of 72 thermocouples have failed since the beginning of the irradiation program. The number of failures per capsule through July 6, 1964, are as follows:

<u>Capsule No.</u>	<u>Thermocouple Failures</u>
43-1	2
43-2	0
43-3	1
43-4	4
43-5	0
43-6	5

Three out of the four thermocouple failures in 43-4 occurred during reactor startup for MTR Cycle 183, the initial cycle of irradiation. Those for 43-6 occurred immediately after the capsule was moved the second time for ETR Cycle 60. The other thermocouple failures occurred at random intervals during the irradiations. Even though twelve failures have occurred, there is still at least one thermocouple for monitoring the temperature of each fuel specimen in the capsules still in-pile.

Two capsules were discharged during April 1964, with an estimated fuel burnup of approximately 40 a/o of $^{235}_{92}\text{U}$ by fission. Capsule 43-1, containing Z-10 and C-10 specimens, was removed from the ETR at the end of Cycle 62

TABLE XII

Maximum Temperature of Specimens by Burnup Quadrants*

Specimen	Burnup Quadrant a/o	Burnup at Maximum Temperature		Maximum Temperature, °F	
		a/o	Fissions/cm ³ x10 ²⁰	Surface	Center
Z-10-B	0-10	7.0	0.8	610	3980
Z-10-B	11-20	15.4	1.7	645	4260
Z-10-B	21-30	28.6	3.2	605	3940
Z-10-B	31-40	36.7	4.0	560	3560
Z-22-M	0-10	9.4	2.5	620	4170
Z-22-M	11-20	14.5	3.8	620	4170
Z-22-M	21-30	27.0	7.1	600	4000
Z-22-M	31-40	39.1	10.3	570	3750
Z-22-N	0-10	3.2	0.8	655	4460
Z-22-N	11-20	12.8	3.4	610	4080
Z-22-N	21-30	--	--	--	--
Z-22-N	31-40	38.4	10.1	630	4260
Z-22-O	0-10	8.8	2.3	710	4840
Z-22-O	11-20	18.6	4.9	635	4260
Z-22-O	21-30	25.9	6.8	620	4170
Z-22-O	31-40	39.5	10.4	595	3960
C-25-M	0-10	2.9	1.1	660	3580
C-25-M	11-20	--	--	--	--
C-25-M	21-30	29.2	11.2	515	2680
C-25-M	31-40	--	--	--	--
C-25-O	0-10	--	--	--	--
C-25-O	11-20	14.1	5.4	610	3240
C-25-O	21-30	25.0	9.6	620	3310
C-25-O	31-40	30.2	11.6	570	3000

* Data for the C-10 series are being analyzed.

(April 13, 1964), and capsule 43-4, containing Z-22 and C-25 specimens, was removed from the MTR at the end of Cycle 207 (April 27, 1964). Both capsules were returned to the Battelle Hot Cell Facility for post-irradiation examination.

Estimated burnups of fuel specimens from capsules 43-1 and 43-4 on discharge from the reactor are shown in Table XIII. Burnup analyses are now being performed.

TABLE XIII

Burnup Data

<u>Specimen</u>	<u>Burnup</u>	
	<u>a/o*</u>	<u>Fissions/cm³ x 10²⁰</u>
Z-10-A	40.0	4.4
Z-10-B	44.7	4.9
Z-10-C	44.6	4.9
C-10-A	41.8	6.4
C-10-B	37.8	5.8
Z-22-M	40.4	10.6
Z-22-N	39.8	10.4
Z-22-O	40.8	10.7
C-25-M	38.0	14.5
C-25-O	39.4	15.1

The discharge schedule for the four remaining capsules, based on conditions at the end of ETR Cycle 64, is shown in Figure 34. While this schedule was obtained from burnup rate estimates that are not unduly optimistic, it may still contain some error, since the opportunity for maintaining satisfactorily high burnup rates during the latter stages of these irradiations via capsule relocations into progressively higher flux facilities is becoming increasingly limited for the following reasons:

*Percent Fission Burnup of U-235 Atoms.

TENTATIVE DISCHARGE SCHEDULE FOR CAPSULES (REMAINING) AT
END OF ETR CYCLE 64

1. The choice of facilities in the reactor is narrowed by the flux requirements.
2. The difference in flux levels in neighboring facilities increases as the overall local level increases.

The implication from the second reason is that the latter part of the exposure in a particular facility may be "below par" from the viewpoint of satisfactory burnup rate; relocation, however, may compromise the specimen-temperature criteria.

C. Post-Irradiation Examination

Post-irradiation evaluation for this program includes the following items:

1. Visual inspection of capsules, specimens within each capsule, and fuel pellets in each specimen.
2. Opening capsules and recovering the specimens and neutron dosimeter wires.
3. Measurement of dimensions, masses, and densities of specimens.
4. Gamma-scanning specimens.
5. Analysis of gaseous fission fragments released from the fuels (subsequent diffusion of same through clad of specimens).
6. Neutron activation analysis of dosimeter wires.
7. Optical metallography of fuel and clad combinations.
8. Measurement of fuel pellet dimensions and densities of fuel materials.

9. Mass spectrometric burnup analyses.
10. X-ray diffractometry..
11. Corrosion testing of fuel materials.

This report contains results of items 1 through 6 and partial data for items 7 and 8. A complete analyses of all data will be provided in an interim report which will be prepared after the hot cell evaluations of these samples have been completed.

1. Visual Examinations and Sectioning of Capsules

The exterior of the capsules, both of which were intact, retained a bright metallic appearance except for a darkened surface area opposite the heat transfer fins of the inner shell.

The outer shell of both capsules was dry-cut at the top and bottom to expose the sodium heat transfer medium to reaction with ethyl alcohol. The specimen trains containing the five fuel specimens and dosimeter wires were recovered intact from both capsules. Typical appearances of ZrO_2 -base and CeO_2 -base specimens are shown in Figures 35 through 38. Slight bowing of the specimens at the lower magnification is an optical effect due to the angle of the camera. The dosimeter wires were subsequently sectioned to lengths corresponding to the specimen lengths for burnup determinations.

2. Dimensional and Density Determinations of Clad Samples

The fuel specimens were lightly wire-brushed to remove adhering particles from the sodium-alcohol reaction prior to dimensional and weight measurements. Micrometer measurements were taken of the outer diameter of each specimen at about one-quarter inch intervals for the 0 and 90

HC 14564

ACTUAL SIZE

APPEARANCE OF IRRADIATED Z-10-B SPECIMEN

Fig 35

HC 14565

6 X

CLOSE-UP OF PORTION OF Z-10-B SPECIMEN

Fig. 36

HC 14568

ACTUAL SIZE

*APPEARANCE OF IRRADIATED C-25-M
SPECIMEN*

Fig. 37

HC 14569

6X

CLOSE-UP OF PORTION OF C-25-M SPECIMEN

Fig. 38

degree positions. Specimen lengths were recorded as an average of three micrometer measurements. Specimen densities were determined by the carbon tetrachloride immersion technique using a Bureau of Standards control.

Pre-irradiation dimensions and pre- and post-irradiation density measurements of the specimens are presented in Tables XIV and XV. Comparisons of these data are shown in Table XVI. The diametral changes for all specimens were less than 0.25%, within the experimental error of measurement. Similarly, the densities of the clad samples decreased by less than 0.5%. These changes are not considered to be significant and are all within or close to experimental error. The root mean square error is 0.0005 inch for geometrical measurements and 0.03 gm/cm^3 for density determinations.

3. Gamma Scans

All clad samples in both capsules were gamma-scanned. The gamma scan curves obtained will be used to determine the burnup of each irradiation specimen and to establish the axial variation in burnup. Mass spectrometric burnup analyses are to be performed at one axial position from one zirconium base fuel pellet in each capsule. Using this data point for normalizing, it will be possible to establish the burnups for the other samples through an analysis of the gamma scan data.

The full length of each specimen was scanned on a calibrated gamma-ray scintillation spectrometer utilizing an eighth-inch diameter collimator. Each specimen was scanned twice with the scanning direction reversed on the second scan. Scans were made at the following spectrometer settings:

TABLE XIVPre-Irradiation Measurements of Clad Fuel Specimens

<u>Specimen Number</u>	<u>Diametral (inch)</u>		<u>Length (inch)</u>
Z-10-A	0.4110	0.4108	2.2520
	0.4111	0.4111	2.2523
	0.4110	0.4111	2.2521
C-10-A	0.3603	0.3604	2.2530
	0.3600	0.3603	2.2531
	0.3598	0.3601	2.2531
Z-10-B	0.4116	0.4111	2.2701
	0.4110	0.4111	2.2699
	0.4107	0.4111	2.2700
Z-10-C	0.4107	0.4108	2.2641
	0.4108	0.4110	2.2637
	0.4104	0.4104	2.2642
C-10-B	0.3601	0.3598	2.2604
	0.3603	0.3600	2.2603
	0.3601	0.3602	2.2602
Z-22-M	0.3109	0.3106	2.5010
	0.3107	0.3104	2.5008
	0.3102	0.3098	2.5008
C-25-M	0.2506	0.2506	2.4980
	0.2506	0.2504	2.4981
	0.2503	0.2505	2.4980
Z-22-N	0.3105	0.3101	2.5003
	0.3100	0.3100	2.5003
	0.3094	0.3096	2.5004
Z-22-O	0.3112	0.3113	2.4984
	0.3113	0.3115	2.4976
	0.3103	0.3103	2.4982
C-25-O	0.2505	0.2507	2.5005
	0.2505	0.2505	2.5005
	0.2504	0.2505	2.5008

TABLE XV

Pre- and Post-Irradiation Densities of Clad Fuel Specimens

<u>Specimen Number</u>	<u>Density (gm/cm³)</u>	
	<u>Pre-Irradiation</u>	<u>Post-Irradiation</u>
Z-10-A	5.63	5.61
	5.63	5.61
	5.63	5.61
C-10-A	6.64	6.64
	6.64	6.64
	6.64	6.64
Z-10-B	5.65	5.62
	5.65	5.62
	5.65	5.62
Z-10-C	5.64	5.61
	5.64	5.61
	5.64	5.61
C-10-B	6.64	6.63
	6.64	6.63
	6.64	6.63
Z-22-M	5.72	5.70
	5.72	5.71
	5.72	5.70
C-25-M	6.58	6.57
	6.58	6.57
	6.58	6.57
Z-22-N	5.73	5.71
	5.73	5.71
	5.73	5.71
Z-22-O	5.72	5.71
	5.72	5.71
	5.72	5.71
C-25-O	(a)	6.68
		6.68
		6.68

(a) Believed to be in error by immersion method. Density by weight and dimensions is 6.70 gm/cm³.

TABLE XVI

Dimensional and Density Changes for Clad Fuel Specimens

Specimen Number	Average Diameter (inch)		Diameter Change (%)	Specimen Density (gm/cm ³)		Density Change (%)
	Pre-Irradiation	Post-Irradiation		Pre-Irradiation	Post-Irradiation	
Z-10-A	0.4110	0.4119	+0.22	5.63	5.61	-0.36
Z-10-B	0.4111	0.4116	+0.12	5.65	5.62	-0.53
Z-10-C	0.4107	0.4117	+0.24	5.64	5.61	-0.53
C-10-A	0.3602	0.3598	-0.11	6.64	6.64	--
C-10-B	0.3610	0.3595	-0.17	6.64	6.63	-0.15
Z-22-M	0.3104	0.3107	+0.10	5.72	5.70	-0.35
Z-22-N	0.3101	0.3105	+0.13	5.73	5.71	-0.35
Z-22-O	0.3110	0.3112	+0.06	5.72	5.71	-0.17
C-25-M	0.2505	0.2506	+0.04	6.58	6.57	-0.15
C-25-O	0.2505	0.2504	-0.04	6.70(a)	6.68	-0.30

(a) Determined from weight and dimensional measurements. Density by immersion was 7.14 gm/cm³ and is believed to be in error. All other densities are by immersion technique.

- a. 0.757 Mev, corresponding to $^{95}_{40}\text{Zr}$.
- b. 0.769 Mev, corresponding to $^{95}_{41}\text{Nb}$.
- c. 0.498 Mev, corresponding to $^{103}_{44}\text{Ru}$.

On the basis of gamma scan data, it was concluded that Z-10-B and Z-10-C were the highest burnup samples in capsule 43-1, whereas Z-22-M and Z-22-O were the highest burnup samples in capsule 43-4. These observations are in agreement with estimated burnups based on temperature gradient data shown in Table I.

4. Burnup Analyses

Samples for burnup analyses have been prepared. Results are not yet available. Mass spectrometric burnup data will be obtained from the following relationship which represents a reasonable approximation of burnup for fuels of high enrichment in which greater than 10% of the fissionable uranium has been consumed.

$$F_5 = \left[\frac{1}{\bar{R}_{5/8}} \right] \left[(\bar{R}_{5/8} - R_{5/8}) - (R_{6/8} - \bar{R}_{6/8}) \right]$$

where, F_5 = a/o of $^{235}_{92}\text{U}$ burnup due to fission of $^{235}_{92}\text{U}$.

$\bar{R}_{5/8}$ = isotopic ratio of $^{235}_{92}\text{U}/^{238}_{92}\text{U}$ in unirradiated fuel.

$R_{5/8}$ = isotopic ratio of $^{235}_{92}\text{U}/^{238}_{92}\text{U}$ in irradiated fuel.

$\bar{R}_{6/8}$ = isotopic ratio of $^{236}_{92}\text{U}/^{238}_{92}\text{U}$ in unirradiated fuel.

$R_{6/8}$ = isotopic ratio of $^{236}_{92}\text{U}/^{238}_{92}\text{U}$ in irradiated fuel.

The loss of $^{238}_{92}\text{U}$ and $^{236}_{92}\text{U}$ is assumed to be negligible.

Data obtained from thermal neutron activation analyses of Ni-0.6 wt% Co dosimeter wires could not be accurately correlated with estimated burnups from thermal gradient data. A comparison of these data are shown in Table XVII. It should be noted that the thermal gradient data and gamma scan measurements exhibited good correlation.

TABLE XVII

Thermal Neutron Activation Analyses

<u>Dosimeter Wire Opposite Given Specimen</u>	<u>n_t vt</u>	<u>Estimated Burnups by Thermal Gradient Data, a/o of $^{235}_{92}\text{U}$ by Fission</u>
Z-10-A	9.9×10^{20}	40.0
C-10-A	9.3×10^{20}	41.8
Z-10-B	1.0×10^{21}	44.7
Z-10-C	1.0×10^{21}	44.6
C-10-B	1.0×10^{21}	37.8
Z-22-M	9.6×10^{20}	40.4
C-25-M	1.0×10^{21}	38.0
Z-22-N	1.1×10^{21}	39.8
Z-22-Θ	7.8×10^{20}	40.8
C-25-Θ	1.1×10^{21}	39.4

5. Fission Gas Release

Sampling for fission gas was performed in two steps. The first consisted of sampling the capsule to determine whether gross sample failures occurred. The second step was associated with measurements from clad samples. Sampling for fission gas was conducted by placing the capsule or fuel sample in a vacuum tight system and drilling a hole through the side wall of the capsule or cladding of the fuel sample. The released gas was transferred by means of a Toepler pump to a gas collection system of known volume. Vials on the gas collection system were sealed and analyzed by either gamma spectrometry (for gas collected from the capsule bodies) or mass spectrometry (for gas collected from the fuel samples).

Data presented in Table XVIII indicate that the quantity of fission gas within the capsules was very low and probably is a result of diffusion through the sample cladding. These low concentrations further substantiate previous conclusions concerning the structural integrity of the samples.

Data for gas release from the fuel samples are presented in Table XIX. The Z-10 and C-10 series samples were irradiated to approximately the same fission density. The lower gas release in the C-10 series is probably a result of a lower relative* temperature of operation than that for the Z-10 series. Although it may appear that the higher gas release in the Z-22 series is a result of the higher fission density, data in Table XII indicate that the Z-22-O sample ran hotter over an extended period of time than did the Z-10-B sample. Z-22-M, which had a thermal history (Table XII)

* Relative to the melting point.

TABLE XVIII

Fission Gas Release to Capsule

<u>Capsule Number</u>	<u>Percent of Total ⁸⁵₃₆ Kr Released to Capsule</u>
43-1	4×10^{-5}
43-4	2×10^{-5}

TABLE XIX

Fission Gas Release From Fuel Samples

<u>Specimen Number</u>	<u>Burnup (Fissions/cm³) x 10²⁰</u>	<u>Fission Gas Release (% Xenon)</u>
Z-10-B	4.9	43
Z-10-C	4.9	32
C-10-A	6.4	22
C-10-B	5.8	28
Z-22-M	10.6	46
Z-22-O	10.7	58
C-25-M	14.5	74
C-25-O	15.1	79

somewhat analogous to, but slightly higher than that of sample Z-10-B, exhibited only a small increase in fission gas release. This observation is consistent with the substantially higher gas release in sample Z-22-O which ran hotter than the Z-22-M sample. Thus, the increased fission gas release in the Z-22 series is probably not a result of the increase in fission density. Further correlations with microstructure are shown in Table XX and will be discussed below. The extremely high gas release in the C-25 series samples was unexpected. Although the high fission density (Table XIX) may be a factor in the high fission gas release in this series of samples, these samples, based on thermal data, ran hotter than the C-10 series and would be expected to release more gas.* It is the magnitude of the difference in gas release between the C-10 and C-25 series which is not clearly understood at this time. However, interesting correlations with microstructure are shown in Table XX. If a fission density dependency is operating, the influence of such a factor will become apparent in the evaluation of the next series of capsules which are being irradiated to higher fission burnups in approximately the same temperature ranges.

A correlation between changes in microstructure and fission gas release is presented in Table XX. A comparison was made of the volume percent grain growth obtained from analyses of post-irradiation microstructures and gas release. Although such a comparison should be sensitive to the time at which a particular microstructure was developed, the relatively good correlation of the data does not indicate a time dependency. It would appear from these data that a reasonable approximation of fission gas

* However, there is some indication in the microstructure that the C-10 samples may have run hotter.

release may be obtained from an analysis of post-irradiation microstructure. On this basis, the unusually high fission gas release for the C-25 series samples is consistent with relative values from other samples. A further analysis of microstructure changes indicates that changes in microstructure of the ceria-base series probably occurs at significantly lower temperatures than those in the zirconia-base series. This is reflected in the fission gas release data already discussed.

TABLE XX
Correlations Between Microstructure
and Fission Gas Release

<u>Specimen Number</u>	<u>Measured Gas Release (%)</u>	<u>Estimated Volume Percent Grain Growth</u>
C-25-O	79	63
C-25-M	74	64
Z-22-M	46	52
Z-10-B	43	47
C-10-B	28	34

Further analyses and correlations between fission gas release, temperature, burnup and microstructure will be provided in a subsequent interim report which will be prepared upon completion of all post-irradiation evaluations for the samples in capsules 43-1 and 43-4. At that time it is anticipated that preliminary fission gas release data from capsule 43-2 (to be withdrawn in February 1965) will provide additional information concerning the influence of fission density.

6. Examination of Microstructures

Examination of microstructures was conducted on transverse sections of the clad fuel samples. The specimens were dry cut with a silicon carbide wheel through the midsection of the center pellet. The as-cut surfaces were photographed at a magnification of 6X to show the crack pattern prior to surface preparation. One section of each specimen was mounted in room temperature setting epoxy resin and prepared for microscopic examinations by metallographic techniques involving grinding, polishing and etching. The polished, unetched surfaces were photographed again at a magnification of 6X. Selected regions of the fuel and cladding structures were photographed at magnifications of 100X to 750X. The same regions of the fuels, in some cases, were rephotographed following etching to delineate the structures.

The etching behavior was strongly dependent upon the oxide system (ZrO_2 -base or CeO_2 -base) and upon the UO_2 concentration. Specific etchants utilized in developing the grain structures of the four fuel compositions are included in Table XXI. Photographs of the as-cut and then polished transverse surfaces of the four fuel specimens are shown in Figures 39 through 42. Pellets of both ZrO_2 -base and CeO_2 -base fuels had fragmented into characteristic blocky sections as a result of the radial and circumferential crack patterns. The low magnification photographs indicated that none of the fuels had undergone unusual structural changes typified by evidence of melting and central void formation or by severe fragmentation. The polished sections of the four fuel compositions showed the presence of annular rings. These patterns of light and dark zones are most evident in the Z-22 pellet (Figure 41)

HC15019

AFTER DRY CUTTING

6 X

HC15562

AFTER MOUNTING
AND POLISHING

6 X

MACROGRAPHS OF TRANSVERSE SECTION OF ZIO-B SPECIMEN

HC 15018

AFTER DRY CUTTING

6X

HC 15554

AFTER MOUNTING
AND POLISHING

6X

MACROGRAPHS OF TRANSVERSE SECTION OF C10-A SPECIMEN

HC 15020

AFTER DRY CUTTING

6 X

HC 15561

AFTER MOUNTING
AND POLISHING

6 X

NOTE: OUTER RING IS CROSS SECTION
OF FUEL SAMPLE CONTAINER

MACROGRAPHS OF TRANSVERSE SECTION OF Z22-M SPECIMEN

HC 15021

AFTER DRY CUTTING

6X

HC 15555

AFTER MOUNTING
AND POLISHING

6X

NOTE: OUTER RING IS CROSS SECTION
OF FUEL SAMPLE CONTAINER

MACROGRAPHS OF TRANSVERSE SECTIONS OF C25-M SPECIMEN

TABLE XXI

Fuel Etchants

Z-10 Fuel

40 v/o conc. lactic acid)	36 v/o conc. HNO_3)	
40 v/o conc. HNO_3)	Swab	36 v/o conc. HF) Immersion
20 v/o conc. HF)		28 v/o H_2O_2)

C-10 Fuel

20 v/o NH_4F)	
50 v/o conc. H_2SO_4)	Immersion
30 v/o H_2O_2)	

Z-22 Fuel

Boiling saturated solution of NH_4F) Immersion

C-25 Fuel

20 v/o conc. H_2SO_4)	
80 v/o H_2O_2)	Immersion

Type 347 Stainless Steel Cladding

50 v/o conc. HNO_3)	
25 v/o conc. HCl)	Immersion
25 v/o glycerine)	

where a second unidentified phase, possibly related to fission product migration, may be appearing. Autoradiographs will be taken to verify relative fission product migration.

The microstructures of the four fuels are characterized in greater detail in the following sections.

a. Z-10 Fuels

This fuel composition exhibited more extensive microstructural changes than any of the other fuels. It also appears to have operated at a substantially higher temperature than any of the other fuels, although temperature monitoring data and fission gas release data have indicated otherwise. Diametral traverses of the unetched structures of fuel specimens Z-10-A and Z-10-B are shown in Figures 43 and 44. The etched structure of the same region of the Z-10-B pellet is shown in Figure 45. The structure may be roughly divided into four zones representing four thermal regions each of which had effected specific changes in the fuel structure. Irradiation effects (metallic fission products and fission gases) are considered singly and in combination with the thermal effects upon the changes in structure. Each zone is described in detail below.

(1) Central Zone

The central zone consisted of a discontinuous, highly porous structure which was not delineated in greater detail by the etchants. This region is similar to that sometimes observed in UO_2 at the high temperature side of a columnar growth region and corresponds to the step immediately preceding the formation of a central void. In fact there is some evidence of the initiation of a central void in Figures 43 to 45. The

RADIAL TRAVERSE OF IRRADIATED Z-10-A FUEL PELLET

Fig. 43

Z-10-B ETCHED 100X

RADIAL TRAVERSE OF IRRADIATED Z-10-B FUEL PELLET

Fig. 45

Z-10-B UNETCHED 100X

RADIAL TRAVERSE OF IRRADIATED Z-10-B FUEL PELLET

Fig. 44

individual particles in this central region have undergone considerable densification as a result of exposure at temperatures substantially above the sintering temperature of 1800°C . Melting did not occur and this is substantiated by the in-pile thermal history of fuel specimen Z-10-B (Tables VI and XII) which indicated a maximum fuel centerline temperature of 2355°C (4270°F), approximately 280°C below the melting point of this fuel composition. The fuel fragments exhibited rounded edges and elongated voids which denoted the highly plastic state of the fuel within this zone. The large voids are associated with void migration up the thermal gradient as well as fission gas agglomeration. Fission gas bubbles are also seen in irregular patterns adjacent to the voids. There was no evidence of microstructural changes which could be attributed to phase transformation. This central zone of both pellets was free from a metallic phase which was developed in the cooler regions of the fuel during irradiation.

(2) Inner Middle Zone

This zone consisted entirely of blocky, radially oriented grains which were outlined by grain boundary voids (Figures 43 and 44). The lenticular pores at the hotter end of the grains have been identified with radial grain growth by vapor phase transport and deposition of fuel material from the hotter to the cooler pore surfaces. This "sweeping action" is believed to be the reason for the larger fraction of fission gases released from the Z-10 fuel as compared to the C-10 fuel (Table XIX) which exhibited a less-developed columnar structure.

The largest amounts of a metallic-like phase were observed within this zone (Figures 43 and 44). This metallic phase

exhibited a variety of shapes ranging from irregular and elongated particles within the cooler region of this zone to globular or spheroidal particles within the hotter region. The variety of shapes of these particles is shown in Figures 46 through 51. Although this metallic phase has not been specifically identified, it is probably a U-Zr-Fs* alloy. Specific identification of this alloy and further evaluation of its properties (particularly melting point) may provide an internal temperature meter based on observations of regions of melting (rounded particles) for this metal phase. Some unusual features of this phase are illustrated in Figure 48 where elongated particles have bridged a crack without fracture indicating a metallic-like plasticity. Figure 49 shows a fine granular structure for this metallic-like phase. Evidence of melting of this phase is shown by the presence of globular particles in the photomicrographs of Figures 50 and 51. A comparison of Figures 44 and 45 indicates that this phase was removed as a result of etching. It was not established whether the particles were dissolved by the etchants or merely removed from the matrix by dissolution at the particle-fuel interface. Additional features of the metallic-like phase will be presented in the descriptions of microstructures of the other fuels.

(3) Outer Middle Zone

This zone is poorly defined in structure and occupies a narrow region between the large grained inner-middle zone and the essentially unchanged outer zone. The limits of the zone may be seen in Figures 43 and 44 within which densification had occurred (with the accompanying reduction in porosity) and within which an apparent second phase has developed (Figure 45). The identity of this phase has not been established

* Fs = Alloying constituents corresponding to fission products with high yields.

UNETCHED

500X

*IRREGULARLY SHAPED METALLIC PARTICLES WITHIN
OUTER ZONE OF Z10A FUEL PELLETT*

Fig. 46

UNETCHED

500X

*METALLIC PARTICLES WITHIN OUTER ZONE OF Z10B
FUEL PELLETT*

Fig. 47

UNETCHED

500X

*ELONGATED METALLIC PARTICLES BRIDGED ACROSS CRACK
LOCATED IN OUTER REGION OF ZIRC FUEL PELLETT*

Fig. 48

ETCHED

750X

*GRANULAR STRUCTURE OF METALLIC PARTICLE IN ZIRC
FUEL PELLETT*

Fig. 49

UNETCHED

500X

*GLOBULAR APPEARANCE OF METALLIC PARTICLES
NEAR CENTER OF FUEL PELLETT*

Fig. 50

UNETCHED

500X

*GLOBULAR METALLIC PARTICLES NEAR CENTER OF
ZIRC FUEL PELLETT*

Fig. 51

at the present time. Furthermore, due to the poor etching characteristics of the structures within this zone, well defined grain boundaries and details of this phase are not delineated. However, it is believed that this apparent second phase is associated with fission product migration. The metallic-like particles within this zone are irregular in shape and are not observed following etching.

(4) Outer Zone

This zone includes the outermost region of the fuel pellets which is in contact with the cladding. Grain structure, pore size and distribution are identical to those for the as-sintered fuel pellets. Due to the low temperatures within the zone, fission gas bubbles would be expected to be of extremely small size with little tendency for agglomeration into larger pores.

An unusual microstructural effect was observed within the portion of the pellets immediately adjacent to the cladding. The appearance of filament-like crack patterns are evident in the unetched structures of Figure 44 and in etched structures of Figures 45, 52 and 53. Both the filament-like cracks and normal cracks near the cladding are filled with a gray deposit which appear to have extended from the fuel-cladding interface (Figure 54). The etching characteristics indicate that the deposits are not epoxy mounting material. A shallow reaction zone has formed at the interface and is believed to be a metal oxide, possibly iron and chromium oxides. Figure 54 also shows this gray material within some of the pores which interconnected with the cracks. This reaction product, which was also present in the ceria-base samples, has not yet been identified.

ETCHED

100X

*CRACK LIKE PATTERN IN PORTION OF ZrO₂ FUEL PELLET
ADJACENT TO CLADDING*

Fig. 52

ETCHED

100 X

*ANOTHER LOCATION OF ZrO₂ FUEL PELLET ADJACENT
TO CLADDING*

Fig. 53

UNETCHED

500X

*APPEARANCE OF GRAY MATERIAL WITHIN
LARGE CRACKS, FILAMENT-LIKE CRACKS
AND IN PORES*

b. Z-22 Fuel

Microstructural evaluations were performed on a single pellet from the center of the Z-22-M fuel specimen. Four distinct zones of microstructural changes, observed in the Z-10 pellet, are seen in the diametral traverse of Figure 55. Etched structures of the outer portion of the Z-22 pellet are shown in Figure 56. Selected areas of significant microstructures within these zones are included in Figures 57, 58 and 59.

(1) Central Zone and Inner Middle Zone

The innermost portion of the Z-22 pellet contained a central core defined by concentric discontinuous void formations, shown in Figure 55 (unetched structure) and 59 (etched structure). This central region exhibited less evidence of void concentration than the same region of the Z-10 pellets, indicating an apparent lower operating temperature than that for the Z-10 series, in contradiction to thermal data. The grain boundaries are delineated by pores in a manner resembling that for the Z-10 pellets. Since the pores are very closely spaced or interconnected, little additional detail of the grain boundaries was developed by etching. However, those portions of the grains which are delineated by etching show very fine boundaries which exhibit no evidence of minor phases with the exception of the metallic-like structure (see middle portion of Figure 58). The quantity and size of the lenticular pores are considerably smaller than those in the Z-10 pellets. It is significant that the numerous metallic-like particles appear in globular form within a region included by about 50% of the diametral volume of the pellet. The location of many particles of this phase within pores at the grain boundaries may be seen in the photomicrograph

DIAMETRAL TRAVERSE OF IRRADIATED
Z-22-M FUEL PELLETT

HC 15186-15187

ETCHED.

100 X

*MICROSTRUCTURE IN OUTER PORTION OF IRRADIATED
Z-22-M FUEL PELLETT*

ETCHED

OUTER
REGION

250X

ETCHED

MIDDLE
REGION

250X

*MICROSTRUCTURES OF THE OUTER AND MIDDLE REGIONS
OF IRRADIATED Z-22-M FUEL PELLETS*

*MICROSTRUCTURE OF A REGION NEAR THE CENTER
OF THE IRRADIATED Z-22-M FUEL PELLETT*

HC 15188

ETCHED

100 X

*MICROSTRUCTURE IN CENTER OF
IRRADIATED Z-22-M FUEL PELLETT*

of the middle region of Figure 57. The observations of less severe mechanical damage, smaller voids and lenticular pores suggests that this fuel was operated at lower fuel centerline temperatures than the Z-10 fuel.

(2) Outer Middle Zone

The third zone, as previously, includes a relatively narrow, highly densified region containing a second phase (Figures 55, 56 and 57) which is delineated in greater detail than in the case of the Z-10 fuel pellets. The cored appearance and complete absence of grain structure suggest a region of highly segregated constituents typical of a cast material. It is probable that this region represents the outer periphery of fission product migration. This possibility will be evaluated by means of autoradiographs. The outer boundary of this zone is defined by circumferential cracks seen in Figure 55.

(3) Outer Zone

The outermost zone of the Z-22 pellet very closely resembles the appearance of the Z-10 pellets, and contains the as-sintered structures, filament-like cracks and a gray phase which may be a result of clad-fuel interaction. The gray material within cracks and interconnected pores is shown in Figure 60.

c. C-10 Fuel

The center pellets from C-10-A and C-10-B specimens were examined for microstructural changes. Radial traverses of unetched structures of the two samples are included in Figures 61 and 62. A diametral traverse of the C-10-B pellet structure in the etched condition is shown in Figure 63.

A comparison of the structures of Figures 61 and 62 indicates that the C-10-A sample had operated at higher temperatures than the C-10-B samples, consistent with thermal data. The central void in Figure 61, although similar in appearance to central voids in irradiated UO_2 , does not appear to be formed by a similar mechanism. The specific mechanism responsible for this void is not understood at this time, especially in light of the microstructure surrounding the void. Melting has been discounted as a possible void formation mechanism on the basis of an analysis of the microstructure surrounding this central void. There is no evidence of a grossly columnar, highly densified microstructure. The etched structure in Figure 64 shows a duplex grain structure containing small equiaxed grains along with large grains oriented in the radial direction.

Large voids exhibit globular to elongated shapes within the highly plastic region of the fuel near the central void. The voids are randomly dispersed within grains or precipitated at grain boundaries (Figure 64). The central region of the fuel appears to have undergone considerable plastic flow which may have resulted in agglomeration of large voids and may have influenced release of the fission gas. The high plasticity of the fuel is also evidenced by the almost complete absence of fine cracks within the central region.

Similar structural changes in grain size and orientation, fission gas void formation and fuel plasticity are noted to a lesser degree within the central portion of the C-10-B fuel sample. (This observation is in agreement with thermal and fission gas release data.) The magnitude of these changes may be compared in Figure 64 and the central portion of

UNETCHED

500X

*APPEARANCE OF GRAY CONSTITUENT
IN CRACKS AND PORES*

RADIAL TRAVERSE OF IRRADIATED C-10-B FUEL PELLET

Fig. 62

ETCHED 100 X

DIAMETRAL TRAVERSE OF IRRADIATED
C-10-B FUEL PELLET

Fig. 63

HC 15034

ETCHED

100X

*MICROSTRUCTURE IN CENTRAL REGION
OF C-10-A FUEL PELLETT*

Figure 63. This region is devoid of the metallic-like phase which developed within the cooler region of the fuel.

The remaining portions of both samples exhibit essentially identical structures. The middle region consists of large grains which are radially oriented within the hotter portion and equiaxed within the cooler portion of the region. Void size decreases, with the majority of the voids occupying random positions within the grains. There are considerably fewer voids at the grain boundaries and essentially complete absence of lenticular voids, as noted for the ZrO_2 -base fuels. The boundaries of the grains are sharply delineated and appeared to be free from minor phases (Figure 63). Trace amounts of the metallic-like phase is observed within this region of the pellets (Figure 65). This phase was not removed completely as a result of etching.

The outer portion of the fuel showed progressively decreasing grain size which approached the as-sintered grain size within the outer region of the pellets. There is some evidence of apparent fission product migration in this region. However, this is not as sharply delineated as in the zirconia-base series. No significant changes in structure are observed for this region of the fuel. A shallow cladding-fuel reaction zone developed probably by the formation of oxides of the cladding material. There is no indication of the filament-like cracks nor of the unidentified gray material within those cracks which extended to the surface of the pellets.

HC 15031

UNETCHED

500X

*METALLIC-LIKE PHASE NEAR CENTER
OF C-10-A FUEL PELLETT*

d. C-25 Fuel

The structure of the center pellets from the C-25-M and C-25-O samples is shown in the traverses of Figures 66, 67 and 68.

The appearance of this fuel is similar to the C-10 series with the exception that the region surrounding the central void appears to have operated at a lower* temperature on the basis of the smaller grain size and lack of evidence of columnar grains. The presence of the central void is not completely understood at this time, especially in view of the apparent low temperature microstructure surrounding the void.

Vapor phase transport is a possible mechanism responsible for central void formation in these fuels on the basis that previous sintering investigations indicated some volatilization at temperatures as low as 1500°C. Continued loss of fuel by volatilization would then enlarge the void into a smooth surfaced axial cavity, shown in Figures 66 and 67. The fuel was transported axially and also deposited within radial cracks extending from the central void. Deposition of fuel within some of the cracks may be the cause for the partial sealing and the resulting irregular width of cracks shown in Figures 66 and 67. The irregular-shaped central void in the two C-10 fuel pellets may have formed in a similar fashion by vapor phase transport.

The middle region of both pellets exhibits equiaxed structures shown in Figure 69. The grains within both the center and middle regions show an irregular appearance resembling the grain boundary markings developed in the thermal stability pellets of the CeO₂-base composi-

* It should be noted that the thermal data from the capsules as well as fission gas release data suggest an opposite conclusion.

*RADIAL TRAVERSE OF IRRADIATED
C-25-M FUEL PELLETT*

DIAMETRAL TRAVERSE OF IRRADIATED C-25-O FUEL PELLET

*RADIAL TRAVERSE OF IRRADIATED
C-25-O FUEL PELLETT*

HC 15142

ETCHED

MIDDLE REGION

500X

HC 15143

ETCHED

NEAR CENTER

500X

MICROSTRUCTURES OF SELECTED REGIONS OF IRRADIATED C-25-M FUEL PELLET

tions (Figure 28). Grain boundary sweeping of fission gases could account, in part, for the large release for this fuel. The middle region contains relatively large amounts of the metallic-like phase (Figures 67 and 70) which appeared to be inert to the etchants (Figure 69). The nature of the gray globular particles seen in the photomicrograph of the central region of Figure 69 is not known.

A third zone of a possible second phase is apparent in the etched structure of Figure 68. The appearance of the structure within this band is not fully developed, however, it is suspected that the same mechanism is responsible for this structure in both the ZrO_2 -base and CeO_2 -base fuels. It is hypothesized that this phase, in all fuels, may be the segregation of fission products at cooler zones of the fuels.

The outer region of both pellets remains essentially unchanged in grain size and in pore volume. The duplex nature of the cladding-fuel reaction zone is shown in Figure 71. The resulting larger volume of reaction products filled the cladding-fuel gap as well as a shallow fissure or crack in the pellet. Filament-like cracks and the presence of a gray material within cracks is not observed in the C-25 fuel.

Figure 71 suggests that the fuel-clad reaction may occur at the expense of the cladding. On the contrary, the reaction may occur at the expense of the fuel, as shown in Figure 72, where the ID of the cladding is smooth while the pellet surface is highly irregular. As noted in Figure 72, the reaction product has occupied the gap space as well as depressions in the pellet surface. This observation provides credence to the hypothesis that the gray material within cracks and pores of the ZrO_2 -base fuels

UNETCHED

500X

*IRREGULAR TO GLOBULAR SHAPED METALLIC-LIKE
PARTICLES WITHIN MIDDLE REGION OF C-25-O FUEL PELLETT*

Fig. 70

*DUPLEX STRUCTURE OF REACTION ZONE AT
CLADDING - FUEL PELLETT INTERFACE OF C-25-O
FUEL SPECIMEN*

UNETCHED

500X

*REACTION PRODUCTS IN GAP BETWEEN
CLADDING AND C-25-M FUEL PELLETT*

is probably the fuel-cladding reaction products.

It should be noted that the apparent fuel-clad reaction layer is no greater than 0.0007 inch. Problems in performance are not anticipated as a result of this apparent reaction layer. Figure 73 illustrates the unchanged austenitic structure of a typical clad sample after irradiation.

In the analyses of the microstructure of these fuels, an anomaly is apparent. For example, thermal and fission gas release data indicate that the Z-22 series operated hotter than the Z-10 series. A similar relationship was established for the ceria-base samples. However, metallographic evaluation of the central region of the fuel indicates that the Z-10 and C-10 series operated hotter than the Z-22 and C-25 series. However, an analysis of the extent of grain growth (Table XX) appears to substantiate the thermal data from the capsules. Evaluation of samples from the other capsules (now being irradiated to higher burnups) should provide additional data required to resolve this apparent anomaly.

7. Fuel Density Measurements and Pellet Dimensions

As a result of fuel cracking, it is anticipated that whole pellets cannot be removed for density determinations. However, the transverse cracking patterns seen in Figures 39 through 42 suggest that pie-shaped sections of fuel can be removed. Proper selection of these sections for measurement should provide density values representative of whole pellets. As a result of previous experience with UO_2 , UC and other ceramic fuels, it is likely that interconnected porosity could lead to inaccuracies in fuel density measurements through use of the conventional carbon

ETCHED

100X

*MICROSTRUCTURE OF TYPE 347 STAINLESS STEEL
CLADDING. (PORTION OF FUEL PELLET IS SEEN
AT BOTTOM OF PHOTOMICROGRAPH)*

tetrachloride methods. On this basis, a technique which utilizes mercury as the immersion medium is being evaluated using unirradiated samples. Mercury is not expected to penetrate pores as readily as carbon tetrachloride.

Work related to fuel dimensional data is still in progress. Available data are presented in Table XXII and are based upon measurements made on metallographic samples. The accuracy associated with these measurements has not been established; on this basis, these values must be considered as preliminary in nature.

The effort on density and dimensions is in progress and further results and analyses will be reported in the next interim report which will be prepared after the hot cell evaluations for these two capsules have been completed.

TABLE XXII

Fuel Pellet Dimensions

<u>Sample Number</u>	<u>Diameter (inch)</u>		<u>Diameter Change (%)</u>	<u>Length (inch)</u>		<u>Length Change (%)</u>
	<u>Pre-</u>	<u>Post-</u>		<u>Pre-</u>	<u>Post-</u>	
Z-10-B	0.3514	0.3521	+0.20	0.4999	0.5042	0.86
Z-10-C	0.3514	0.3505*	-0.25	0.4992	0.5046	1.08
C-10-A	0.3005	0.3032	+0.90	0.5009	0.5083	1.47
Z-22-M	0.2501	0.2505	+0.16	0.4993	--**	--**
C-25-M	0.1894	0.1890*	-0.21	0.3604	--**	--**

* Probably within experimental error.

** Measurements not completed.

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