NASA CR-72282
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TOPICAL REPORT

HIGH TEMPERATURE COMPATABILITY OF UO_2 WITH W-25 Wt % Re

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R. A. Ekvall and J. O. Hegland

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NATIONAL AERONAUTICS AND SPACE ADMINISTRATION

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Technical Management
NASA Lewis Research Center
Cleveland, Ohio
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HIGH TEMPERATURE COMPATIBILITY OF UO_2 WITH W-25 Wt % Re

By R. A. Ekvall and J. O. Hegland

ABSTRACT

The high temperature compatibility of two types of clad fuels, UO_2 contained in tungsten-25 wt % rhenium (W-25 wt % Re), and tungsten-60 and 70 vol % UO_2 cermets (W-60 vol % and 70 vol %) contained in W-25 wt % Re was investigated. Samples of stoichiometric UO_2 clad with W-25 wt % Re were tested isothermally in cesium (Cs) vapor of 1 to 5 torr at temperatures of 1800 to 2200°C for times up to 5000 hours. In addition, some samples of stoichiometric UO_2 clad with W-25 wt % Re were subjected to thermal cycling from a maximum temperature of 2200°C to a minimum of 725°C for relatively short times in a vacuum environment.

Cermet fuels of W-60 and 70 vol % UO_2 clad in W-25 wt % Re were tested in a vacuum ($\sim 10^{-6}$ torr) at 1800 and 2000°C for times to 500 hours.

The results of the evaluation of the compatibility as determined by metallographic analysis, electron microprobe, emission spectroscopy, and wet chemistry are presented.

SUMMARY

Work performed under Contract NAS 3-2544, was directed toward the determination of the high-temperature chemical compatibility between UO_2 and W-25 wt % cladding. This consisted of two parts:

Part 1. Isothermal testing of vented and unvented UO_2 clad in W-25 wt % Re in a Cs environment for times to 5000 hours at temperatures from 1800 to 2200°C. In addition, thermal cycling test of similar materials

were performed in a vacuum environment.

Part 2. Testing of unvented W-UO_2 cermets containing 60 and 70 vol % UO_2 and contained in W-25 wt % Re at temperatures of 1800 to 2000°C for times of 500 and 389 hours respectively in a vacuum environment.

Under Part 1, W-25 wt % Re containers of approximately 0.50-inch diameter by 0.50-inch long with a 0.020-inch thick wall and a 0.030-inch thick end cap were used. A 0.020-inch diameter vent hole was located on one end cap for the vented capsules. Uranium dioxide pellets of stoichiometric composition were placed in the containers and tungsten inert gas welding (TIG) was used to join the end caps to the tube section. The test samples were enclosed in a tantalum (Ta) test chanber which contained a Cs atmosphere. This test chamber was inserted in a vacuum furnace for the isothermal testing.

Capsules isothermally tested for times of 5000 hours at 1800°C, 3000 hours at 2000°C and 1000 hours at 2200°C changed very little in dimension and weight. The visual observations of the capsules revealed no surface defects and no evidence of attack by the Cs vapor. The vent holes were not plugged, although a few vents contained an internal peripheral metallic deposit.

The Cs vapor was very effective in suppressing the evaporation loss of UO_2 through the vent hole relative to the UO_2 loss into vacuum.

In all the capsules examined there was no evidence of gross interaction between the UO_2 and cladding although evidence of a superficial interaction on the inside surface of the cladding was observed in the capsules tested at 2200°C. This interaction zone was approximately 1 μ thick and consisted of a fine grain boundary penetration of UO_2 and a small amount of a metallic phase that may be free U. Varying degrees of porosity was observed in the cladding and was more pronounced for the capsules tested for long times and

at high temperatures. Abnormal grain growth occurred in the cladding of some of the capsules. Spectrographic and chemical analysis of the cladding indicated that a small pickup in O (55 to 100 ppm), U (48 to 360 ppm), and Ta (~ 900 ppm - probably from the chamber) had occurred.

As a result of the isothermal testing, the UO_2 pellets exhibited grain growth. The largest grain size was observed in the high temperature (2200°C) capsule. In addition, free U particles were detected. The origin of the U particles is related to the formation of substoichiometric UO_{2-x} at high temperatures; upon subsequent cooling to room temperatures, free U is precipitated as fine particles randomly dispersed in UO_2. A nonmetallic phase observed in the UO_2 was identified as alumina (Al_2O_3).

In Part 2, cermet samples of 60 and 70 vol % UO_2 in W were inserted into containers which were structurally and chemically identical to those used in Part 1. Bonding of the cermet to the cladding was achieved by hot isostatic treatments at 1650°C in 10,000 psi He.

The clad cermets were tested at 1800 and 2000°C for 500 and 389 hours, respectively in a vacuum environment. The cermets were to be tested for times to 1000 hours at both test temperatures.

However, testing of all the samples was terminated prematurely; extensive container cracking occurred in all samples. All dimensions of the samples increased; the lengths from 0.4 to 2.4% and diameters from 0.6 to 2.4%. Metallographic observations revealed that the W-25 wt % Re cladding was no longer bonded to the cermet and that UO_2 evaporated through the cracked region in the cladding.

The reason for the failures is believed to be related to the large difference in the thermal coefficient of expansion between the cermet and the cladding. The linear thermal expansion of the W-60 vol % UO_2 cermet is approximately 30% greater than the W-25 wt % Re cladding at 1800 to 2000°C.

Microprobe analysis revealed essentially no change in composition of the cladding.

INTRODUCTION

For some space power systems, nuclear fuel materials clad in a suitable material will be required to operate at high temperatures (1600°C and higher) for prolonged periods of the order of 10,000 hours. Under these operating conditions, the fuel material must be chemically compatible with its cladding to assure reliability, i.e., mechanical and thermal integrity.

The chemical compatibility of UO_2 with W and Mo has been reported.[1,2] Since W-25 wt % Re has good high temperature creep properties, ductility, and fabricability superior to that of both W and Mo, a program was initiated to determine the chemical compatibility of W-25 wt % Re with UO_2.

This report is divided into two parts; in Part 1, compatibility studies of W-25 wt % Re and UO_2 are described; in Part 2, testing of W-UO_2 cermets clad with W-25 wt % Re is discussed.

PART 1: TESTING OF UO_2 WITH W-25 WT % Re to 2200°C

A. Isothermal Test of Vented and Unvented Capsules in Cesium Environment

In Part 1, the compatibility studies performed during the early phase of this program, up to 2000 hours of testing, are reported. After this work was completed, a programmatic decision was made to extend the testing times to 5000 hours and to include a test temperature of 2200°C (Phase II). The results of the Phase I tests are summarized while the Phase II tests are discussed in detail.

Experimental Procedures

Sample Geometry - A right cylinder was selected for the specimen configuration. The UO_2 pellets were fabricated with dimensions that would provide sufficient room for thermal expansion of the UO_2 to contact the cladding wall at the designed test temperatures. Vented and unvented samples were prepared, which were identical except for a 0.020-inch diameter vent hole in one cap at each of the vented samples. Figure 1 shows a cutaway view of a typical sample. Dimensions of pellets and containers were:

Pellet (length)	~ 0.22 in.
Container length, L, Figure 1	~ 0.4 in.
Container outside diameter, D, Figure 1	~ 0.45 in.
Container (wall thickness)	~ 0.020 in.
End cap (thickness)	~ 0.030 in.
End cap (weld preparation lip height)	~ 0.030 in.
End cap (weld preparation lip thickness)	~ 0.020 in.

Fuel Fabrication and Characterization - The UO_2 pellets were fabricated as follows:

1. Uranium dioxide powder, containing approximately 3 wt % water as a binder, was compressed under 20,000 psi to obtain the green pellets of approximately 55% of the theoretical density of UO_2.

2. The green pellets were then sintered between 1700 and 1750°C in dry hydrogen for 4 hours. The total time in the furnace was 8 hours including heating and cooling cycles. The sintered pellets were 97 $\pm$ 1% of theoretical density.

3. The sintered pellets were processed to final diameter by centerless grinding and to final length by hand grinding.

4. Before encapsulation, the pellets were heat-treated in hydrogen at 1000°C for several hours to remove surface oxidation which may have occurred during grinding. Table I provides data on the

spectrographic analysis and the ratio of atomic percent oxygen to atomic percent uranium (O:U) of a typical pellet. Figure 2 is a photomicrograph of an as-fabricated UO_2 pellet.

TABLE I
SPECTROGRAPHIC ANALYSIS AND O:U RATIOS OF UO_2

Element	ppm
Ag	< 0.1
Al	250
B	< 0.2
Bi	2
C	3
Ca	25
Cd	< 1
Co	< 2
Cr	50
Cu	5
F	5
Fe	500
Mg	2
Mn	2
Mo	< 3
Na	< 35
Ni	200
Pb	< 1
Si	35
Sn	2
V	15
Zn	< 10
O:U Ratio	2.014

Container Fabrication and Characterization - W-25 wt % Re cladding material was obtained as as-sintered tube and end caps from Chase Brass Corporation; as-extruded tube was obtained from Nuclear Metals Inc. (NMI). Hoskins-sintered W-25 wt % Re rod was used as the end cap material for the NMI tube. The chemical analysis of the W-25 wt % Re material is presented in Table II. Photomicrographs of the as-extruded NMI tube are shown in Figure 3. No sigma phase was observed at magnifications as high as 750X. The W-25 wt % Re tube produced by Chase Brass and Copper Company was

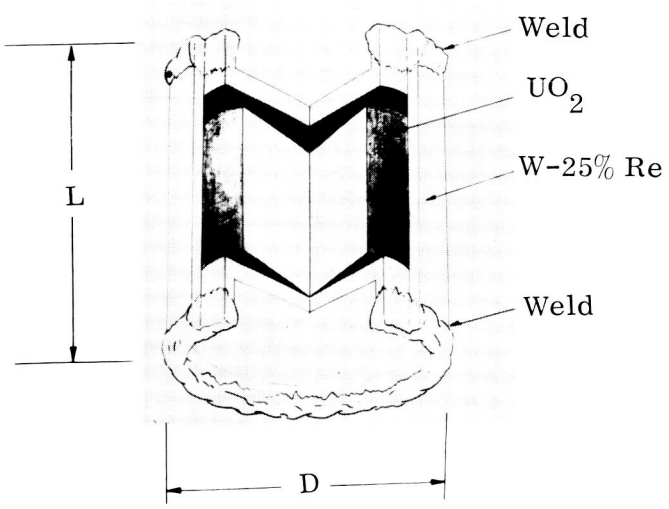

Figure 1. Test Sample Configuration

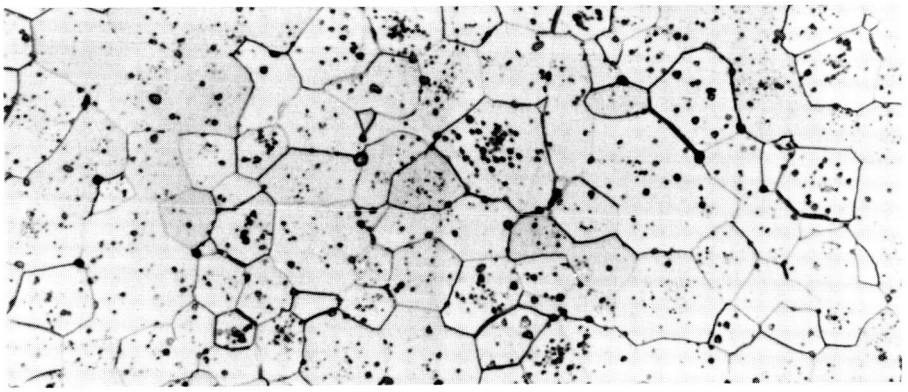

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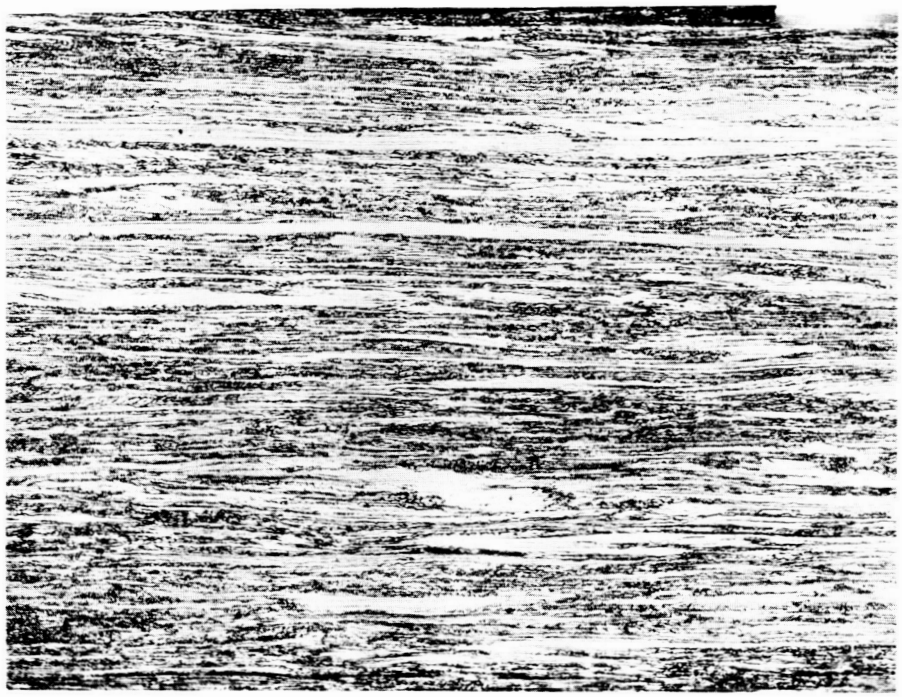
Figure 2. Photomicrograph of UO_2 Pellet



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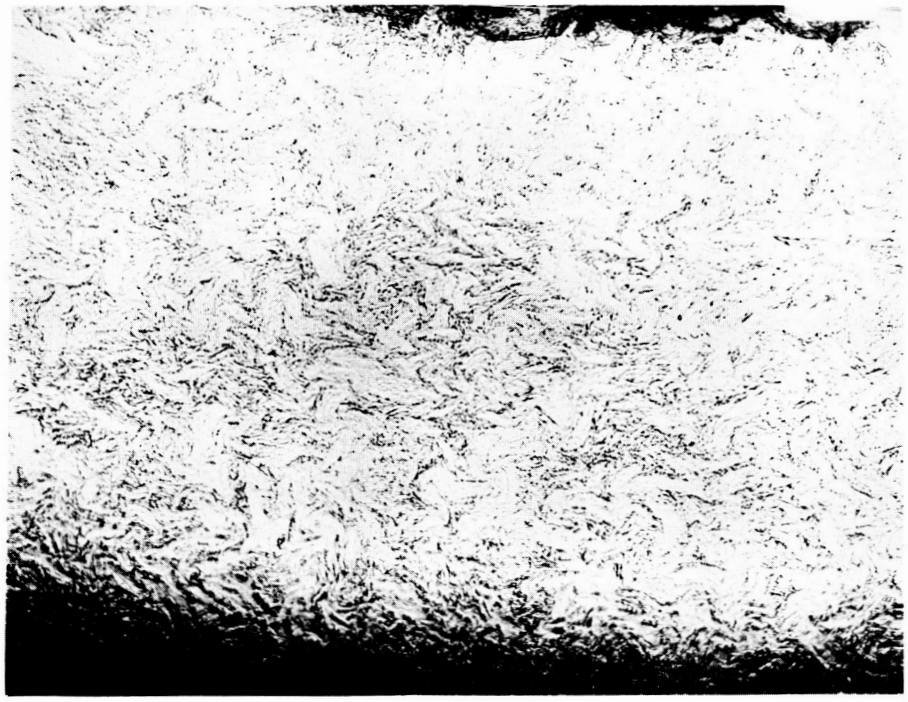
a. Longitudinal Section



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b. Transverse Section

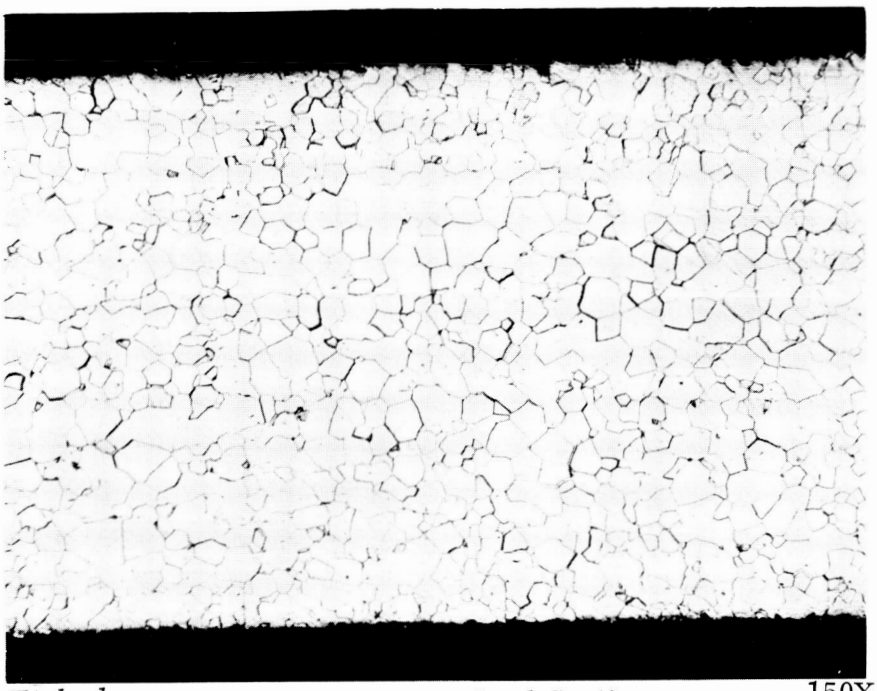
FIGURE 3. PHOTOMICROGRAPHS OF THE AS-EXTRUDED NMI TUBE

examined metallographically. No sigma phase was detected at 500X. Microstructures of the longitudinal and transverse sections are illustrated in Figure 4. A typical microstructure of the Hoskins W-25 wt % Re rod is shown in Figure 5. Chemical and spectrographic analyses of the material were made, and the data obtained are given in Table II. Both the rod and tube material were inspected with fluorescent penetrant (Zyglo); the tube material also was leak checked with a helium mass spectrometer leak detector.

TABLE II

CHEMICAL ANALYSES OF AS-RECEIVED W-26% Re MATERIAL

Element	Hoskins Rod	NMI Tube	Chase Brass Caps	Chase Brass Tube	Analytical Technique
Re	25.0%	24.86%	24.89%	24.95%	Gravimetric precipitation
C	< 10 ppm	32 ppm	26 ppm	31 ppm	Conductometric
O	< 10	60	40	50	Inert Gas Fusion
N	< 10	120	100	100	Micro Kjeldahl
Al	< 10	< 10	< 10	10	Emission spectrographic
Ca		< 1	< 1	< 1	"
Cr	< 10	< 10	< 10	< 10	"
Cd	<100	< 20	< 20	< 20	"
Cb		< 50	< 50	< 50	"
Cu	< 10	10	20	18	"
Cs		< 50	< 50	< 50	"
Fe	< 10	11	7	35	"
Hf	< 50	<100	<100	<100	"
Mo	180	182	115	125	"
Ni	< 10	8	5	10	"
Na		< 5	< 5	< 5	"
Si	< 10	< 20	< 20	< 20	"
Sn		< 20	< 20	< 20	"
Ta	<250	<100	<100	<100	"
Ti	< 10	3	1	2	"
Zr		4	1	3	"
Zn		<100	<100	<100	"

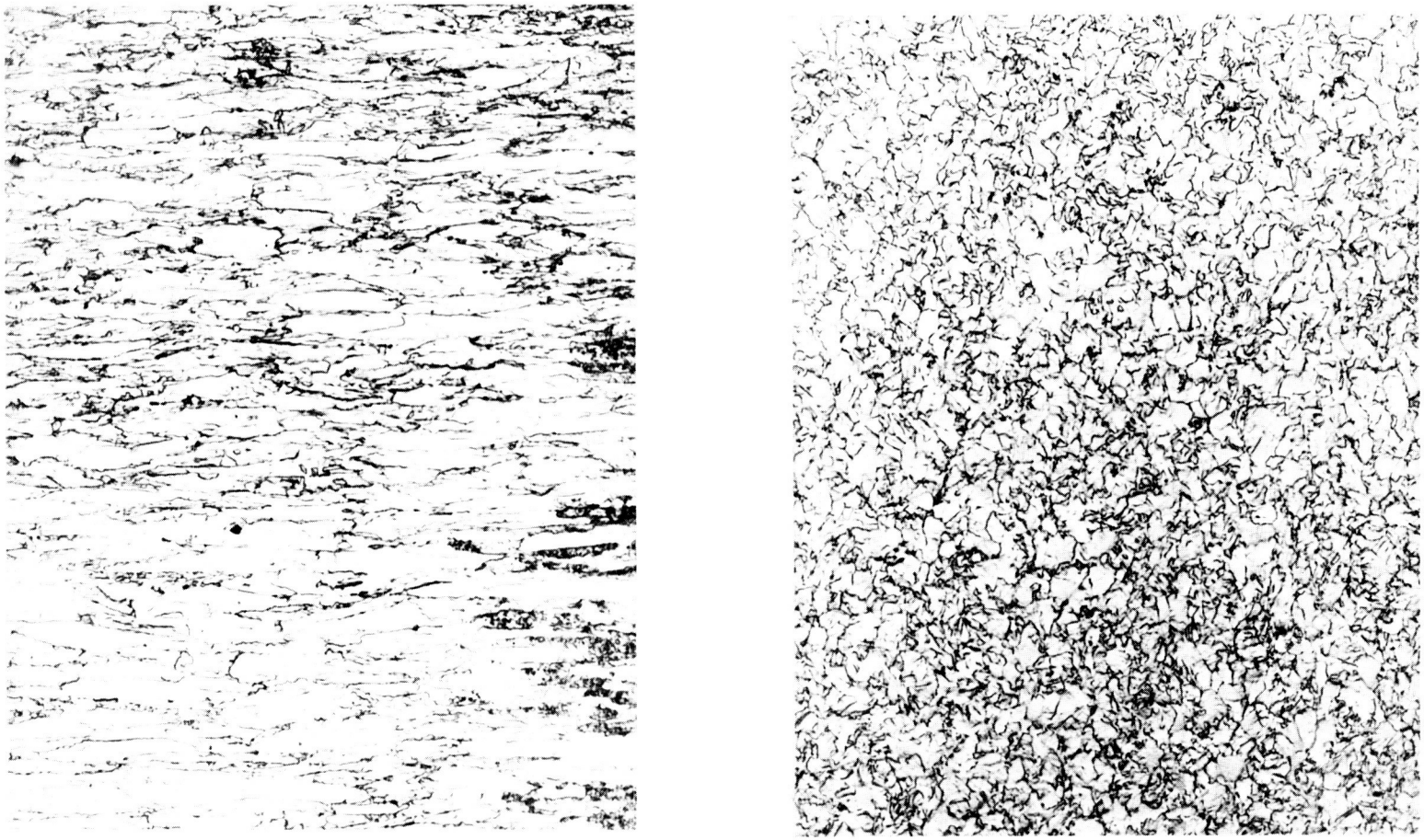


Etched a. Longitudinal Section 150X

Etched 150X

b. Transverse Section

FIGURE 4. W-25 Wt % Re TUBE FROM CHASE BRASS



Etched 100X

a. Longitudinal Section

b. Transverse Section

FIGURE 5. TYPICAL AS-RECEIVED W-25% RE ROD FROM HOSKINS

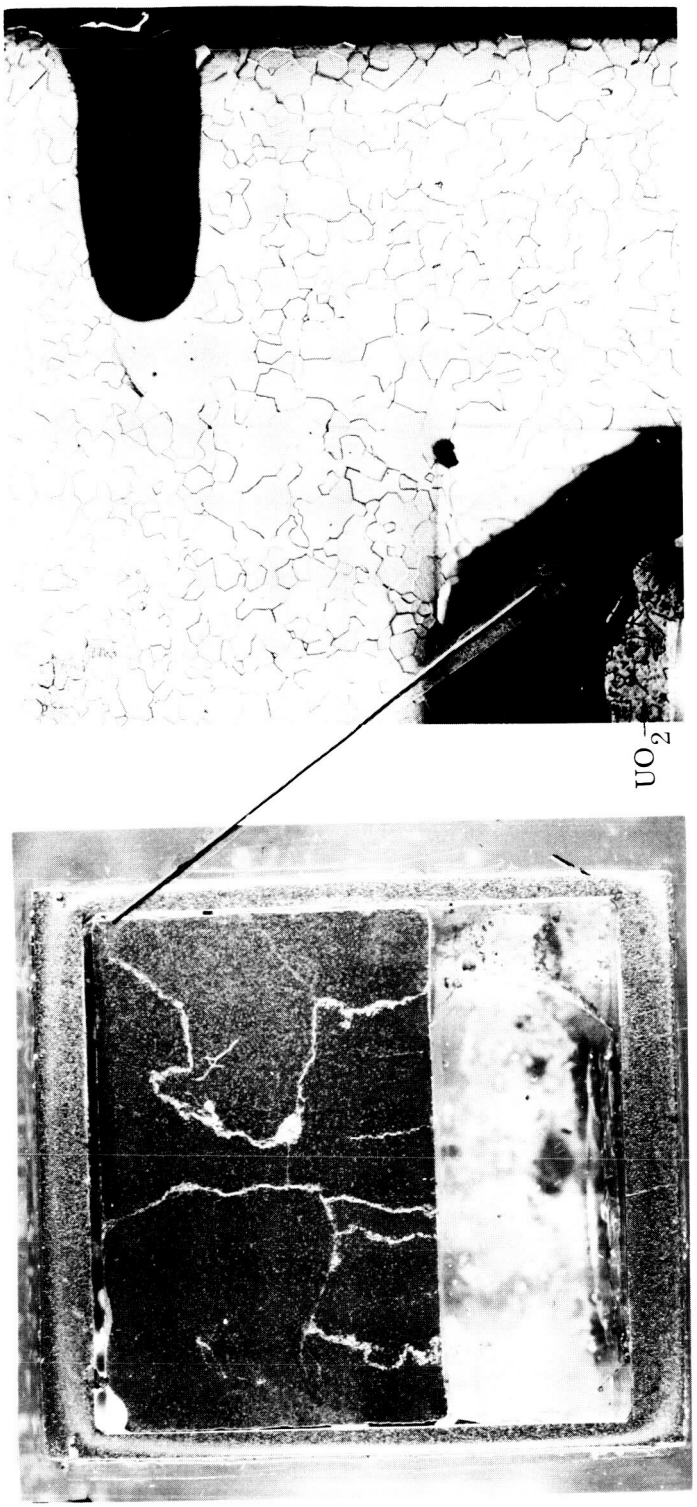
Capsule Assembly Techniques - All but two capsules were fabricated by TIG welding the metal end caps to the tube section. The other two specimens were assembled with a Mo-50 wt % Re braze alloy, melting point approximately 2550°C. This braze was made in a vacuum with an r-f heat source. A macrophotograph of the capsule used in braze experiments and a typical braze microstructure are illustrated in Figure 6.

The welding operation was performed in an argon-filled (< 100 ppm oxygen) glove box, 1/3 to 1/2 atm of argon remained in each container. The material welded readily with better than 90% yield. Figure 7 illustrates typical vented and unvented capsules. Welded capsules were pressurized in a helium bomb and were leak checked with a helium mass spectrometer leak detector to ensure leaktightness. After welding, each capsule was macroscopically examined, and the dimensions and weight were recorded.

Test Procedures

Cesium Environmental Chamber - The test chamber contained the cesium atmosphere to which the capsules were exposed during isothermal testing in the furnace. Figure 8 shows the completed test chamber which was fabricated entirely from tantalum by TIG welding end caps to a tube section to form a chamber approximately 3 inches long, and 1 inch in diameter, with a wall thickness of 0.020 inch. A 1/4-inch diameter tube was welded to each end cap over a hole in the center of the cap. The tube connected to the bottom end cap was seal welded before assembly. The tube attached to the top cap was left unsealed until the remainder of the test components were assembled in an argon atmosphere.

After approximately 1 gm of liquid cesium was poured into the chamber through the tube connected to the top end cap, the chamber was evacuated and then the tube was pinched and seal welded. A spectrographic and oxygen analysis of the cesium was made, and the data obtained are given in Table III. After welding and leak checking, the test chamber was ready for insertion into the vacuum test furnace.



b. Microstructure of Braze Area

a. Macroshot of Capsule

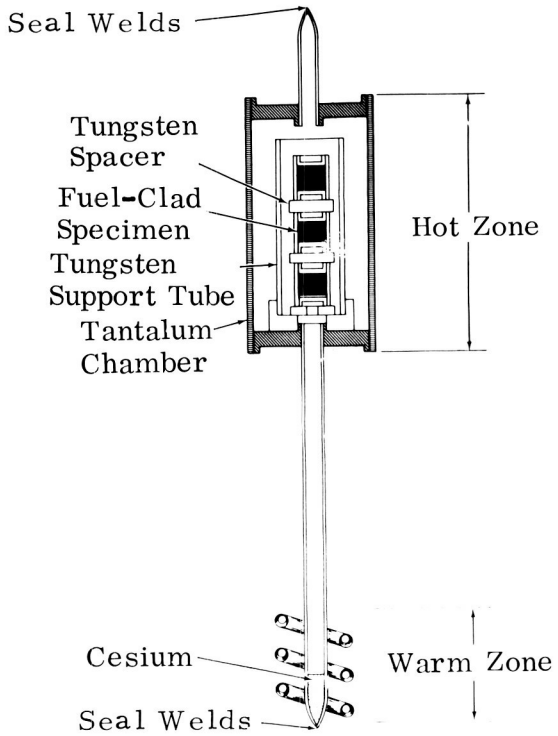
FIGURE 6. AS-BRAZED CAPSULE

a. Vented

b. Unvented

3X

FIGURE 7. TYPICAL CAPSULES AFTER WELDING



a. Illustration of Cesium Test Chamber

b. Test samples and spacers stacked on the bottom cap of test chamber, the tantalum tube section, end cap, and tungsten support tube are shown from left to right in background.

FIGURE 8. CESIUM TEST CHAMBER

TABLE III. CHEMICAL ANALYSIS OF CESIUM

Element*	ppm	Element*	ppm	Element*	ppm
Ag	< 1	Co	< 10	Pb	< 10
Al	10	Cu	3	Sb	< 10
B	< 10	Fe	3	Si	10
Ba	< 10	Mg	1	Sn	< 10
Be	< 1	Mn	1	Si	< 10
Bi	< 10	Mo	< 10	V	< 10
Cd	< 100	Na	< 100	W	< 100
Ca	50	Nb	< 100	Zn	< 50
Cr	1	Ni	< 10	Zr	< 100
		O	400 ± 200		

*All elements were determined spectrographically except oxygen

Vacuum Test Furnaces - A dual, vacuum-furnace unit used in performing the tests consisted of two independently controlled furnaces serviced by a common vacuum system. Each furnace consisted of independently controlled hot and warm zones. The hot zone was housed in a large, cylindrical, stainless steel bell, and the warm zone was accommodated in a smaller, cylindrical, stainless steel housing located directly beneath the hot zone. The two zones were separated by a water-cooled, stainless steel plate with a small hole in the center to permit the long 1/4-inch tube on the bottom of the cesium environmental chamber to extend from the hot zone into the warm zone.

The hot zone heated the test capsules to the desired test temperature. The warm zone provided an independent temperature control for the cesium reservoir. The cesium pressure in the hot zone was regulated by controlling the temperature in the reservoir in the warm zone.

The hot zone was resistively heated by a tungsten, split-ring, coil-type heating element which could provide temperatures up to 2300°C for short periods. The temperature was sensed by a W-5 wt % Re/W-25 wt % Re

thermocouple and was controlled to within ± 1% of the test temperature above 1800°C. The warm zone was resistively heated by a nichrome element designed for a maximum operating temperature of 400°C. The temperature was sensed by a Chromel-Alumel thermocouple and controlled within ± 5°C. Vacuums of 10^{-6} to 10^{-7} torr were maintained in each furnace at all temperatures.

Test Program - The conditions of the isothermal tests are given in Table IV. All capsules received some unavoidable cycling because of utility failures.

TABLE IV

TEST PROGRAM FOR SHORT TERM COMPATIBILITY TESTING OF UO_2 CLAD WITH W-25 WT % Re IN CESIUM (1 to 5 torr)

Phase I - Test Conditions

Capsule	Temperature Tested °C	hours	Vented
A	1800	500	Yes
B	1800	500	No
C	1800	1000	Yes
D	1800	1000	No
E	2000	500	Yes
F	2000	500	No
G	2000	1000	Yes
H	2000	1000	No
I	2000	2000	Yes
J*	1800	1000	Yes
	2000	1000	

*Capsule J was exposed to 1000 hours at 1800°C before being tested at 2000°C.

Phase II - Test Conditions

Capsule Number	Test Temperature (°C)	Test Time (hours)	Vented
1[a]	1800	2000	Yes
2[a]	1800	2000	Yes
3[b]	1800	500	Yes
4[b]	1800	1000	Yes
5	1800	3000	Yes
6	1800	3000	Yes
7	1800	5000	Yes
8	1800	5000	Yes
9	2000	2000	Yes
10	2000	3000	Yes
11	2200	500	Yes
12	2200	1000	Yes

[a] Cladding of all capsules except 1 and 2 was sintered W-25 wt % Re from Chase Brass. The cladding of capsules 1 and 2 consisted of extruded tube from NMI and sintered caps from Hoskins.

[b] All capsules except 3 and 4 were TIG welded. Capsules 3 and 4 were brazed with Mo-50 wt % Re.

Post-Test Evaluation

Inspection and Measurements - After thermal testing, each capsule was Zyglo inspected and macroscopically examined to detect surface defects. The capsules were measured and weighed to determine dimensional and weight changes. Each unvented capsule also was helium leak checked.

Metallography - After inspection, only vented samples were prepared for metallographic examination. For each vented capsule a hole was made in the container wall opposite the vent end to allow for filling with epoxy which maintained the pellet in place in the capsule during sectioning and polishing operations. The capsule was then sectioned longitudinally and mounted in epoxy. Routine metallographic grinding and polishing were used to prepare the capsule for examination on the as-polished condition. Structural details

of the capsule were revealed by etching the UO_2 (for 2 to 3 minutes) with a solution of 90% H_2O_2 and 10% H_2SO_4, and by etching the W-Re (30 to 40 seconds) with a solution of 1 part of 30% potassium ferricyanide and 1 part of 10% NaOH.

Microprobe and Chemical Analysis - Several of the long-term capsules were analyzed by an electron microprobe for evidence of interdiffusion of the elements of the container and UO_2 and identification of inclusions and/or precipitates.

Results and Discussion

Phase I

Vented and unvented capsules were inspected, and the only defect discovered was a small surface pit in capsule A. The unvented capsules were leaktight. The results of Phase I, the short term tests, are summarized below.

Vented capsules showed no gross interaction or interdiffusion of fuel and container elements, or cladding compositional changes after isothermal testing in cesium (1 to 5 torr) at 1800°C (up to 1000 hours) and 2000°C (up to 2000 hours).

Only traces of superficial interaction (e.g., UO_2 inclusions in the container) were observed; these were small, localized, and always in the immediate vicinity of the interface between the container and the UO_2.

Phase II

The results of Phase II, the long term compatibility tests, are presented in greater detail below.

Post-Test Measurements - The post-test measurements of the capsules are presented in Table V. Capsules were visually examined and the surfaces were found free of defects and showed no evidence of attack by the Cs vapor. The vent holes of capsules 7, 8, 9, and 10 appeared to contain an internal peripheral metallic deposit. Further examination of the vent hole will be presented in the discussion of metallographic analysis.

TABLE V. POST-TEST CAPSULE MEASUREMENTS

Test Conditions			Dimensional and Weight Changes		
$T^{o}C$	Hrs	Capsule Number	$\Delta D^{(a)}$ (in.)	$\Delta L^{(b)}$ (in.)	$\Delta W^{(c)}$ (gm.)
1800	2000	1	None	None	-0.0161
1800	2000	2	None	+0.003	-0.0997
1800	500	3	None	None	-0 0033
1800	1000	4	None	None	-0.0052
1800	3000	5	None	None	-0.0187
1800	3000	6	None	None	-0.0172
1800	5000	7	None	None	-0.0229
1800	5000	8	None	-0.005	-0.0195
2000	2000	9	None	+0.002	-0.0059
2000	3000	10	+0.002	None	-0.0098
2200	500	11	None	None	-2.1318[d]
2200	1000	12	+0.002	None	-2.0750[d]

a ΔD = Difference between pre- and post-test capsule diameter. Approximate initial diameter = 0.450 in.

b ΔL = Difference between pre- and post-test capsule length. Approximate initial length = 0.450 in.

c ΔW = Difference between pre- and post-test capsule weight. Approximate initial weight = 13 gm. Aproximate initial UO_2 weight = 6 gm.

d = The Cs chamber ruptured during the first 500-hour exposure and submitted capsules to a vacuum of 10^{-6} to 10^{-7} which increased evaporation by comparison with samples seeing only Cs.

Post-test dimensional measurements revealed no unusual changes. The decrease in length of capsule 8 is believed to be a result of an error in the original capsule measurements.

The weight changes were measured for each capsule and are also presented in Table V. By comparing these results with the calculated weight losses expected for the UO_2 evaporation into a vacuum, [3, 4] the Cs vapor is effective in reducing the evaporation loss of UO_2 by several factors. The large weight losses of capsules 11 and 12 were a result of a rupture in the tantalum chamber during the first 500 hours of testing. The rupture of the chamber permitted the escape of the Cs cover gas and exposed the capsules to the outside vacuum environment which greatly increased the evaporation loss of UO_2. The last 500 hours of thermal testing of capsule 12 were performed in a new Cs containment chamber under a Cs cover gas.

Metallographic Analyses of W-25 wt % Re Cladding and UO_2 Interface - Capsule 2 was sectioned and examined metallographically. A macrograph of the longitudinal section is illustrated in Figure 9a. The fractures in the UO_2 pellet were probably caused by the sectioning operation. Observations of the W-25 wt % Re-UO_2 interface revealed no evidence of interaction. A typical interface is shown in Figure 9b. The separation between the UO_2 and cladding was designed in the capsule to allow for the UO_2 expansion (UO_2 expansion > W-25 wt % Re) in heating the capsule to the test temperature. No evidence of sigma phase was detected in the cladding material. Grain size and hardness measurements were made on the W-25 wt % Re cladding and were essentially unchanged.

Capsules 3 and 4, tested at 1800°C for 500 and 1000 hours, respectively, were examined. These capsules were fabricated by brazing the end caps to the tube section with a Mo-50 wt % Re alloy. There was no evidence of any UO_2/W-25 wt % Re interactions or evidence of sigma phase. The braze areas of the capsules were examined and porosity was observed in these regions. The microstructures are shown in Figure 10. There was no

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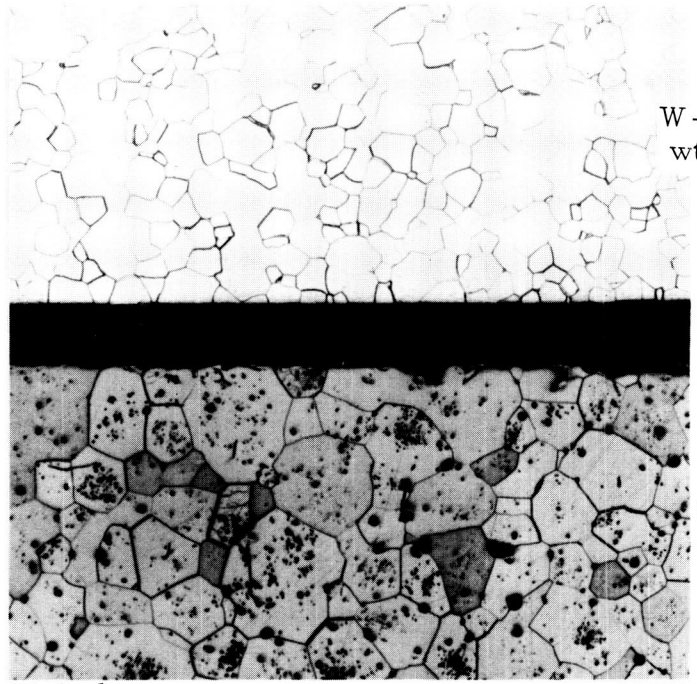
a. Macrograph of longitudinal Section



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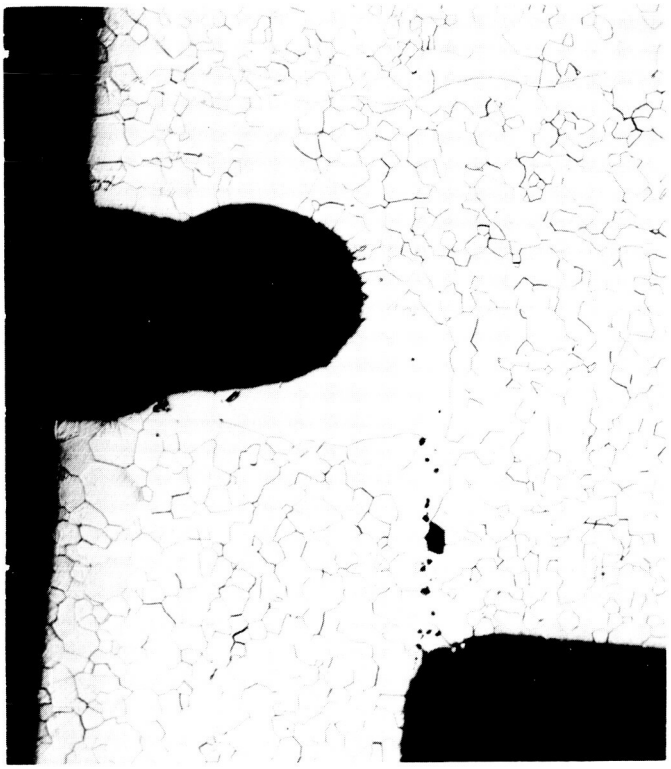
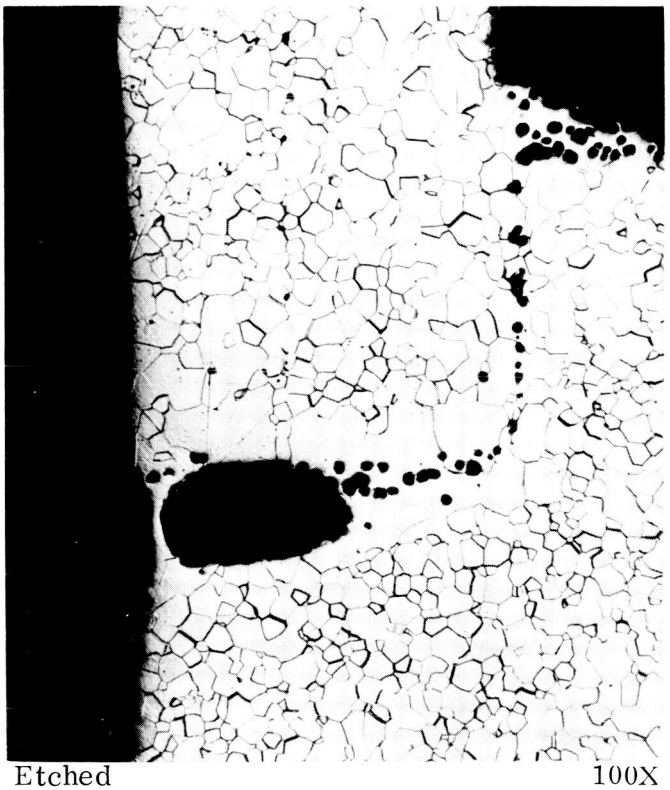
b. Typical Interface Between W-25 wt % Re and UO_2
Longitudinal Section of Container Wall

FIGURE 9. CAPSULE NO. 2 1800°C - 2000 hours



a. Capsule 3 - 500 hours



b. Capsule 4 - 1000 hours

Etched 100X

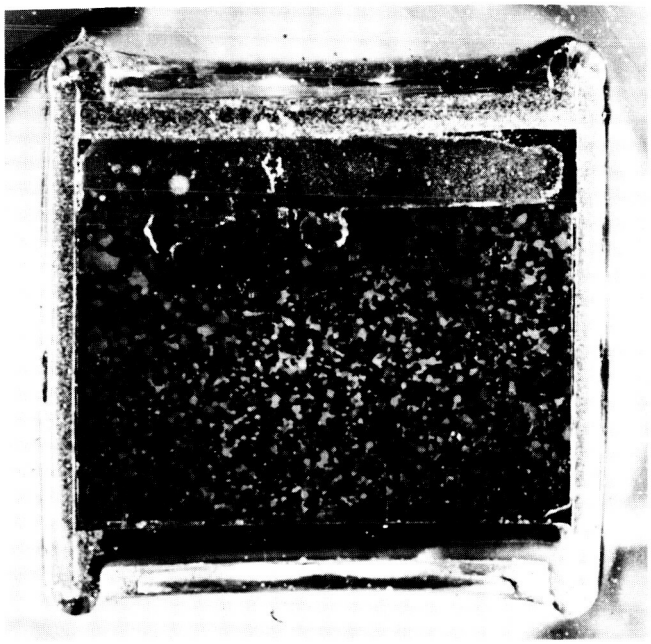
FIGURE 10. BRAZE AREAS IN CAPSULES TESTED AT 1800°C (Longitudinal Section)

porosity in the original braze area, but began to develop along the center of the braze zone after 500 hours at 1800°C and was more pronounced after 1000 hours. The large void in Figure 10b (braze joint interface region) is due to incomplete filling in of the braze joint and is not due to thermal treatment. The formation of porosity is believed to be associated with the Kirkendall effect,[5] i. e., when two metals having different diffusion coefficients are permitted to interdiffuse there is a net transport of material across the plane that initially separated the two. In the brazed capsule, it is believed that Mo diffuses out of the braze area faster than W can diffuse in the opposite direction and a void area is created in the braze.

Hardness measurements were made across the braze areas. In moving from the center of the Mo-50 wt % Re into the cladding, the hardness gradually increased to a maximum value outside the original braze-W-25 wt % Re interface and then decreased to the hardness of the cladding. The hardness maximum is probably associated with a local zone of a hard ternary solid solution.

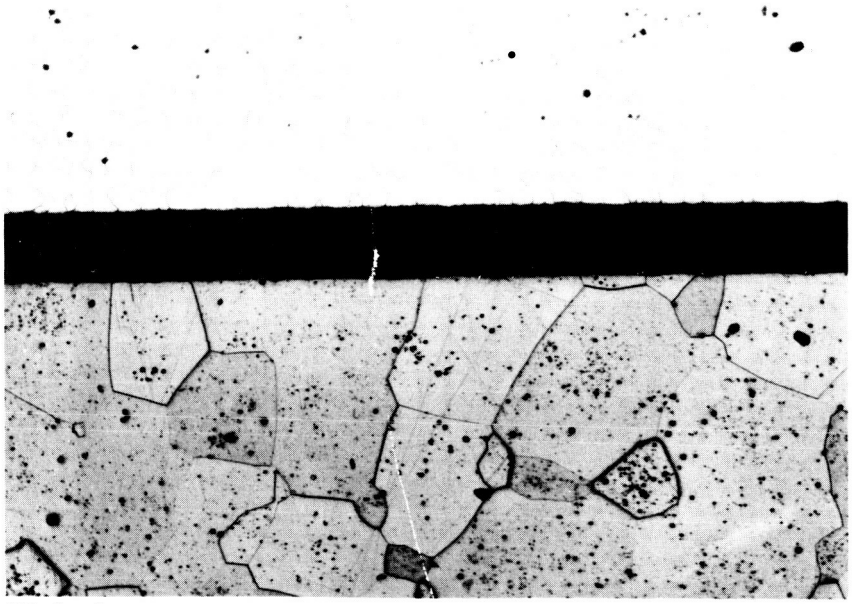
Metallographic observations of capsule 5, 3000 hours at 1800°C, were performed. No interaction between the W-25 wt % Re cladding and the UO_2 was discernible. Further, there was no evidence of sigma formation. Some porosity was observed in the cladding wall and an abnormally large grain was seen in the end cap.

Metallographic analysis of capsule 7, tested for 5000 hours at 1800°C was performed. No evidence of interaction between the UO_2 and W-25 wt % Re was detected. There was an indication that the W-25 wt % Re and UO_2 tend to collect in the vent after prolonged testing times at 1800°C. A macrograph of the longitudinal section of the capsule is shown in Figure 11a, and the microstructure of the UO_2 cladding interface is illustrated in Figure 11b. Porosity was observed in the tube section of the capsule (Figure 11b); however, the end caps contained very little porosity. In one end cap of the capsule, very large grains were observed. The grain size of the clad tube



Etched 6.5X

a. Longitudinal Section of Capsule



Etched 100X

b. W-25 wt % Re/UO_2 Interface

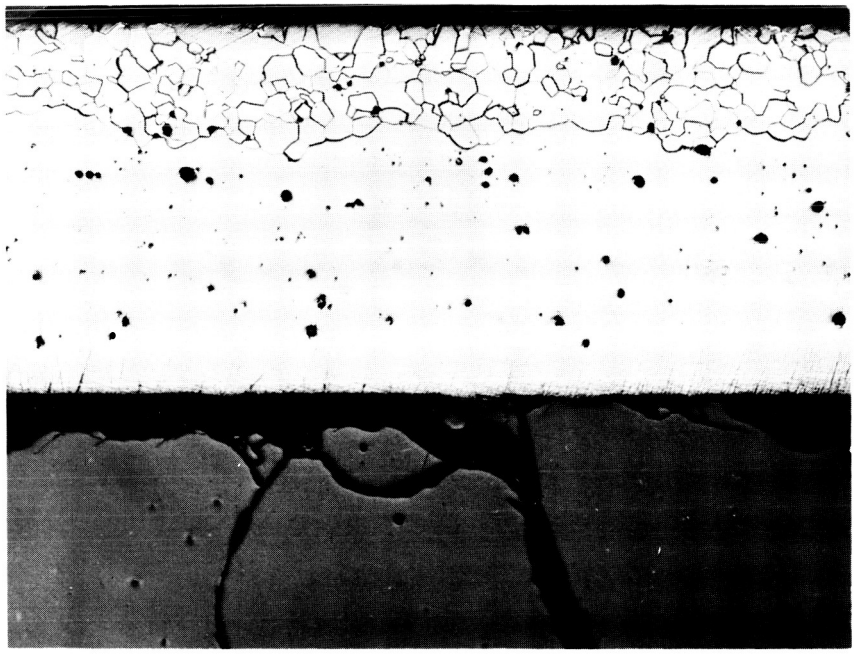
FIGURE 11. CAPSULE NO. 7, TESTED AT 1800°C FOR 5000 HOURS

did not exhibit any change relative to the as-processed tube material.

Examination of capsule 10, 3000 hours at 2000°C, revealed no gross interaction between the UO_2 and W-25 wt % Re cladding. No evidence of sigma phase was detected up to magnification of 500X. A typical interface between the UO_2 and cladding material is shown in Figure 12a. As seen in this figure, abnormal grain growth has occurred in the tube section of the cladding. The material in one end cap had also undergone large grain growth. Porosity was observed in the tube section of the capsule and in the TIG weld area. An example of the porosity in the tube and weld area is shown in the microstructure of Figure 12b. In this capsule, as in capsule 7, there was a metallic deposit found in the vent hole. This will be discussed in more detail in the section on microprobe analysis.

Capsules 11 and 12 were tested at 2200°C for 500 and 1000 hours, respectively. In the course of testing both capsules, the Ta chamber ruptured during the first 500 hours of testing which resulted in a high loss of UO_2. Metallographic observations revealed no gross interaction between the UO_2 and the W-25 wt % Re; typical interfaces of capsules 11 and 12 are shown in Figures 13 a and b, respectively. However, at higher magnifications of 1000X, a thin layer, approximately 1 μ thick of a metallic appearing phase was observed along the inner edge of the nonvented end cap of capsule 11 (Figure 14). This thin layer was attacked by a conventional UO_2 etchant which indicated U. A superficial interaction was also detected at the interface of the UO_2 and the vented W-25 wt % Re end cap of capsule 12. A small amount of phase of the appearance of UO_2 penetrated grain boundaries immediately along the edge of the inside of the cladding as shown in Figure 15. No evidence of this type of superficial interaction was observed in any other capsule.

No sigma phase was observed in the cladding material. The walls and end caps of both capsules 11 and 12 showed very large grains extending in some cases across the entire thickness of the wall or end cap. A much



a. Typical Interface Between UO_2 and W-25 wt % Re

b. Weld Area of Capsule

FIGURE 12. CAPSULE NO. 10, TESTED AT 2000°C FOR 3000 HOURS

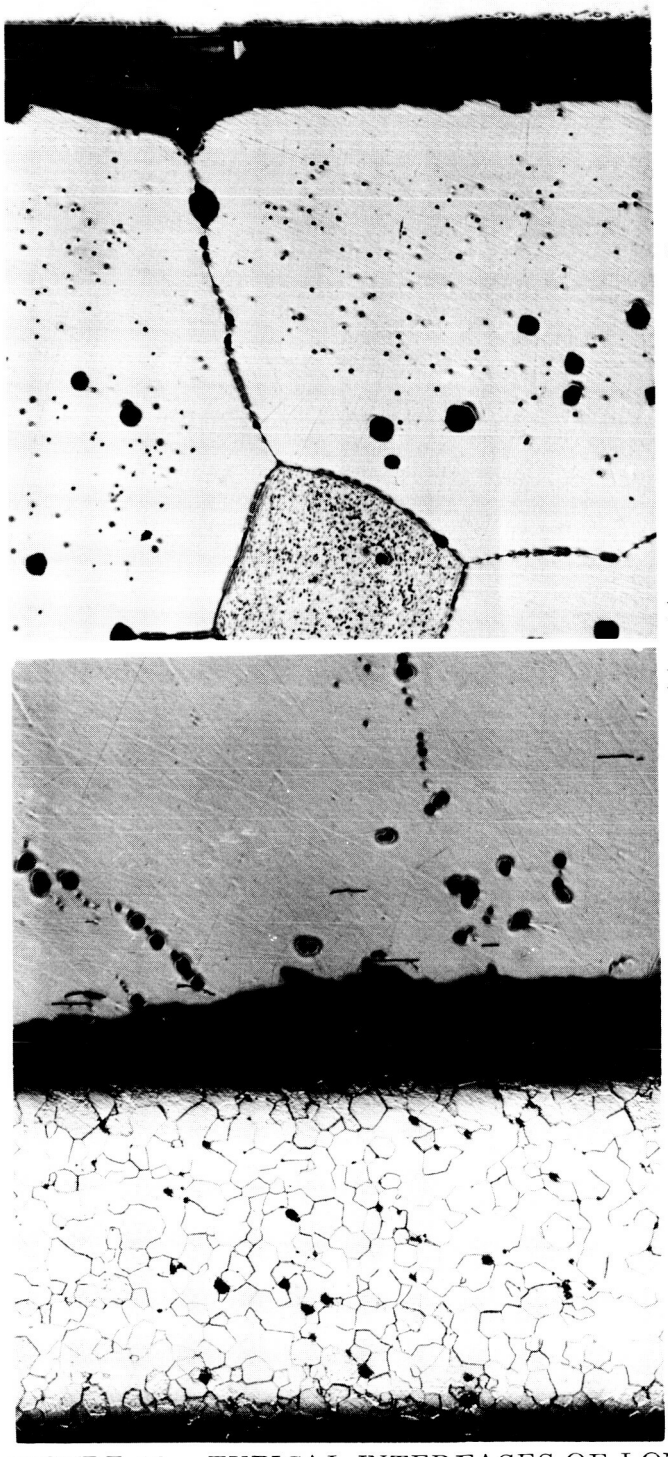


FIGURE 13. TYPICAL INTERFACES OF LONGITUDINAL SECTIONS OF CAPSULES TESTED AT 2200°C

FIGURE 14. THIN LAYER OF METALLIC APPEARING PHASE ALONG THE INTERFACE BETWEEN UO_2 AND END CAP CAPSULE 11 (2200°C - 500 hours)

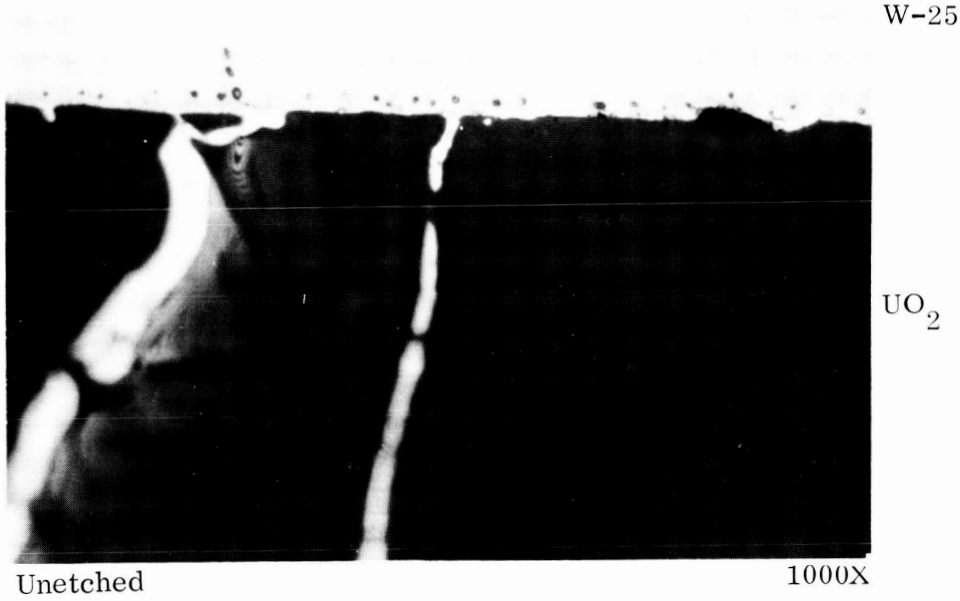


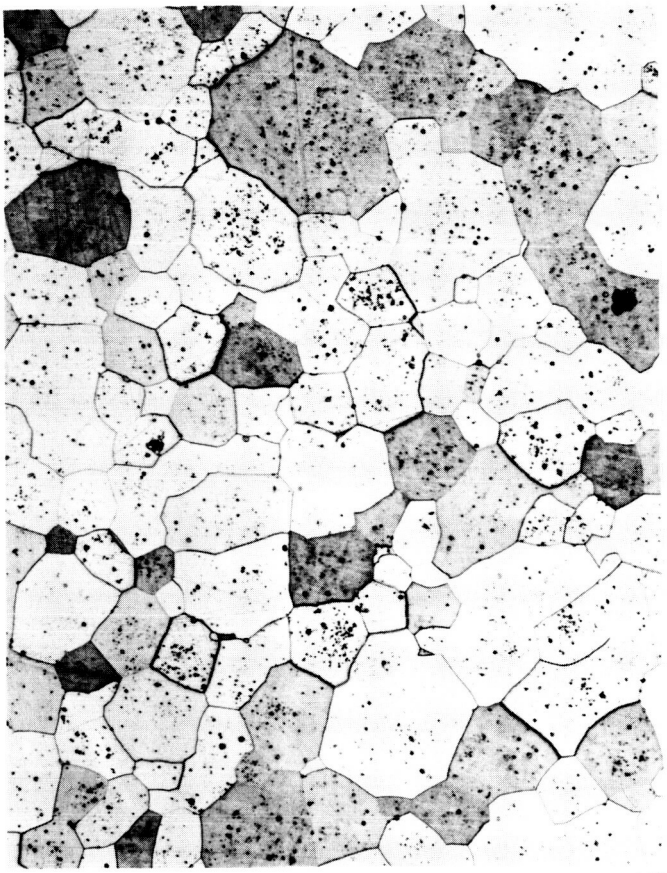
FIGURE 15. NONMETALLIC PHASE (Dark Phase) PENETRATING W-25% Re END CAP GRAIN BOUNDARIES ALONG INTERFACE WITH UO_2. WHITE PHASE IN UO_2 APPEARS TO BE METALLIC. CAPSULE 12. (2200°C - 1000 hours)

smaller equiaxed grain structure existed in the remaining areas. Porosity was observed in the tube section of the capsules, with very few pores in the end cap material. Although metallography of as-received W-25 wt % Re showed no evidence of pores, radiography indicated some light porosity in sections of the tube. It may be, therefore, that the pores observed in the thermally treated samples were associated with pores in the tube before testing. Another explanation may be that micropores present, but not observable, at room temperature coalesced with increasing temperature and time. A third possibility is that the pores developed from the accumulation and resultant pressure of impurities in the W-25 wt % Re which could be gaseous at the high test temperatures.

Metallographic Analysis of UO_2 Fuel - In all capsules examined metallographically, grain growth was evident in the UO_2. This result is to be expected because the test temperatures of 1800, 2000, and 2200°C are higher than the UO_2 sintering temperature of 1700 to 1750°C. The grains in the UO_2 remained equiaxed; a typical example is illustrated in Figure 16. The extent of grain growth is evident by comparing the grain size of the UO_2 of Figure 16 (100X) with those of the as-sintered UO_2 pellets shown in Figure 2 (500X). In the capsules tested at 2200°C, the UO_2 exhibited very large equiaxed grains.

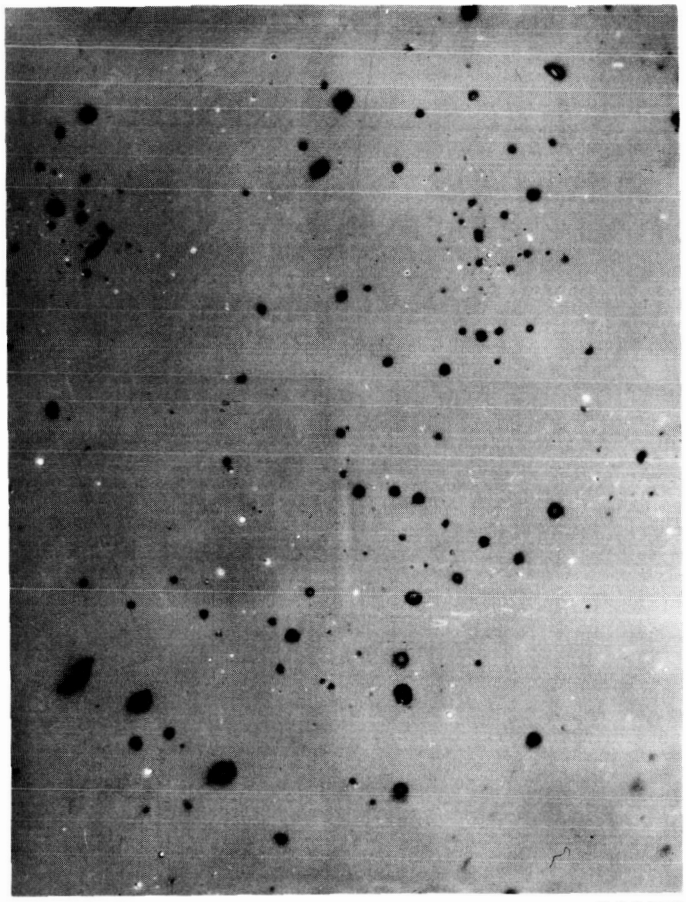
The UO_2 in all capsules contained a metallic appearing phase of varying morphology. In some capsules, e.g., capsules 3 and 4, 500 and 1000 hours at 1800°C respectively, the metallic phase was randomly distributed in very fine particles (Figure 17). In the UO_2 of other capsules, the metallic phase was precipitated in the grain boundaries, pores and cracks as shown in Figure 18. The amount of the metallic phase appeared to increase with an increase in test temperature. (Compare Figures 17 and 18, 1800°C versus 2200°C, respectively.)

The metallic phase appears to be uranium which is probably related to the high temperatures to which the samples were subjected. Experimental evidence[2, 6, 7] has shown that UO_2 becomes substoichiometric (UO_{2-x}) above 1600°C, releases oxygen, and retains the excess uranium in solution



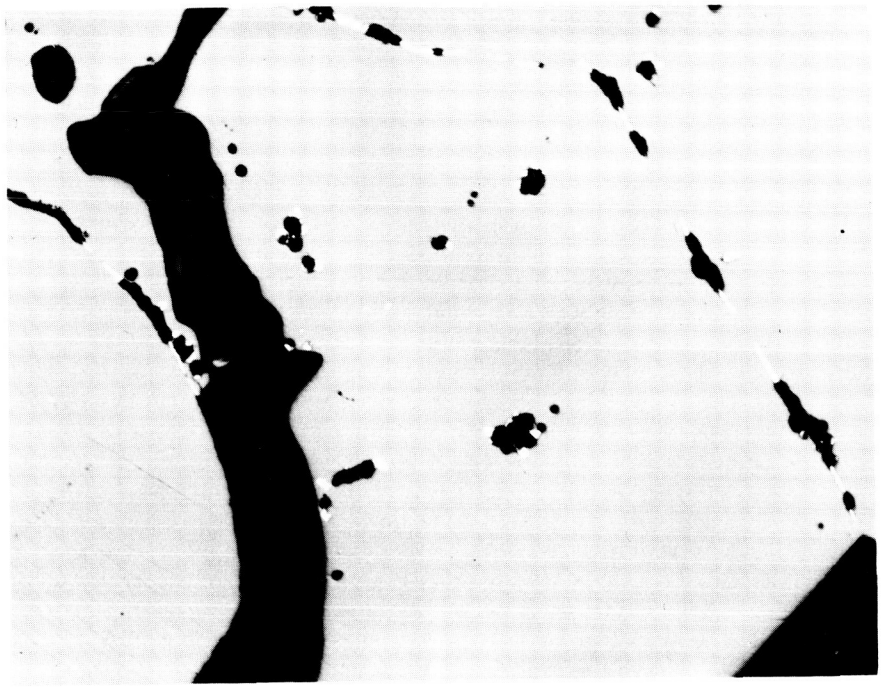
Etched 100X

FIGURE 16. GRAIN STRUCTURES OF URANIUM DIOXIDE OF CAPSULE NO. 7 (1800°C - 5000 hours)



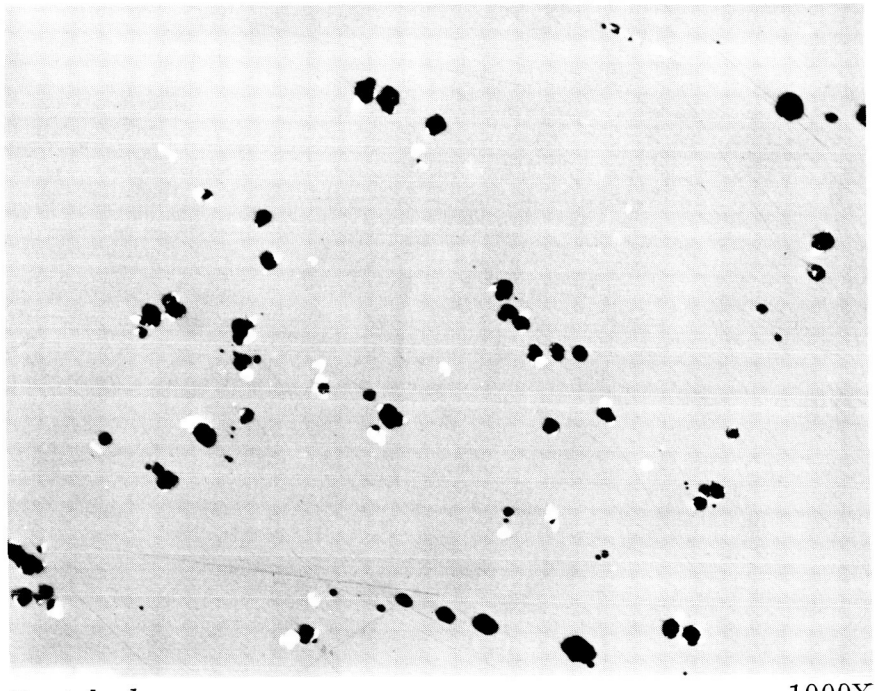
Unetched 1000X

FIGURE 17. METALLIC PHASE TYPICAL OF THAT FOUND IN THE UO_2 OF CAPSULES 3 AND 4, TESTED AT 1800°C FOR 500 and 1000 HOURS, RESPECTIVELY



Unetched 1000X

a. Metallic Phase in UO_2 Grain Boundary of Capsule 11 (2200°C - 500 hours)



Unetched 1000X

b. Metallic Phase in Pores of UO_2 of Capsule 12 (2200°C - 1000 hours)

FIGURE 18. METALLIC PHASE IN UO_2

in the UO_{2-x} structure. The degree of substoichiometry increases with increasing temperature. Upon cooling, the UO_{2-x} reverts to the stoichiometric composition, $UO_{2.00}$ and ejects the excess uranium. Depending upon the rate of cooling, the uranium precipitates in varying amounts within the UO_2 grains and/or at grain boundaries and pores.

In addition to the metallic phase found in the UO_2, a nonmetallic phase was observed in the UO_2 of the capsules tested at 1800 and 2000°C. The nonmetallic phase was not detected in either of the capsules tested at 2200°C. In these capsules, the second phase may have evaporated at these high temperatures. The morphology of the nonmetallic phase was similar to that of the metallic phase, i.e., randomly distributed as fine particles within the grains, concentrated in the grain boundaries of the UO_2 or in cracks. The identification of this nonmetallic phase will be presented in the following section on microprobe analysis.

Microprobe and Chemical Analysis - Microprobe analysis was performed on selected samples in an attempt to identify inclusions and to search for evidence of interdiffusion of the elements of the cladding and UO_2. Relatively large metallic inclusions (up to 20 μ) found in as-sintered UO_2 contained iron, nickel, chromium and silicon, i.e., stainless steel. Two of the inclusions had the following compositions:

	% Iron	% Nickel	% Chromium	% Silicon
1.	78-80	4-5	8-10	5-6
2.	26-27	56-58	9-10	6-7

The very small metallic inclusions (< 1μ) could not be analyzed because of their small size.

The nonmetallic phase observed in as-sintered pellets and in UO_2 after isothermal treatments was probably Al_2O_3. Spectral scans of the inclusions also showed the presence of a small amount of uranium. The uranium indication

probably resulted from x-ray fluorescent excitation of the UO_2 surrounding the Al_2O_3 inclusions.

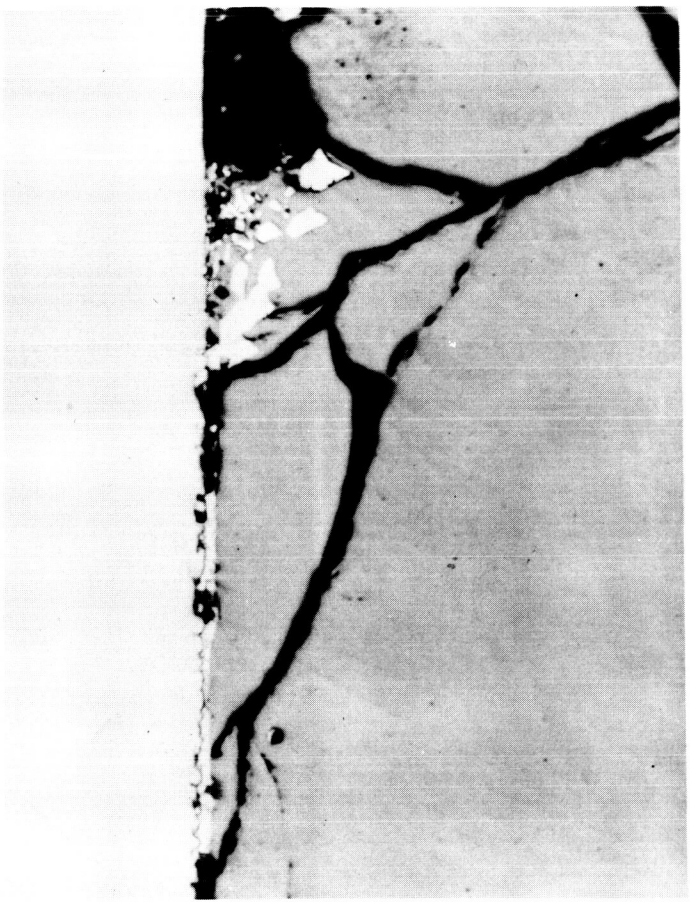
Most metallic inclusions in the UO_2 matrix of capsules No. 11 and 12 (2200°C - 500 and 1000 hours respectively) were 100% uranium. A few inclusions in the immediate vicinity of the cladding cap were found to be uranium with up to 6% rhenium. (Figure 15) Where metallic stringer-type inclusions extended into the UO_2 from the cap, rhenium was found as far back into the UO_2 as 0.16 mm. A depletion in rhenium content was found along the edge of the cap in contact with the UO_2. The rhenium concentration in the cap varied from a nominal concentration of ~ 24.9% rhenium at a distance of 200 μ from the interface to as little as 21.6% about 5 μ from the interface. The very thin band of metallic phase that was found in contact with the nonvented end cap of capsule No. 11 was too narrow to obtain definitive results.

Another type of metallic inclusion not previously observed was found in two areas along the UO_2 end cap interface of capsule No. 11 when it was repolished for microprobe analysis, Figure 19. The microprobe showed these inclusions contained ~ 72% uranium, ~ 13% aluminum, ~ 10% rhenium, and ~ 5% tungsten. The aluminum existed as an impurity in UO_2. No aluminum was found in the UO_2 or clad immediately adjacent to the inclusions.

The nonmetallic inclusions seen in the cap of capsule No. 12 (Figure 15) contained uranium but were too small for quantitative analysis. The uranium content of the particles, their fluorescence in the electron beam and their reddish color under polarized light indicated the particles were UO_2.

On capsule No. 12, several slow scans (10 μ/min) and a number of spot checks of the cladding and UO_2 revealed no aluminum, rhenium, tungsten, or cesium in the UO_2 and no uranium, aluminum, or cesium in the cladding except for the inclusions already noted. The limits of detectability were uranium in tungsten-25% rhenium, ~ 0.16%; aluminum in tungsten-25% rhenium, ~ 0.03%; cesium in tungsten-25% rhenium, ~ 0.02%; tungsten in UO_2, ~ 0.28%;

W-25% Re -



- UO_2

Unetched 500X

FIGURE 19. METALLIC INCLUSIONS IN UO_2 IN CAPSULE NO. 11 (2200°C - 500 hours)

rhenium in UO_2, ~ 0.16%; aluminum in UO_2 ~ 0.04%; cesium in UO_2, ~ 0.02%.

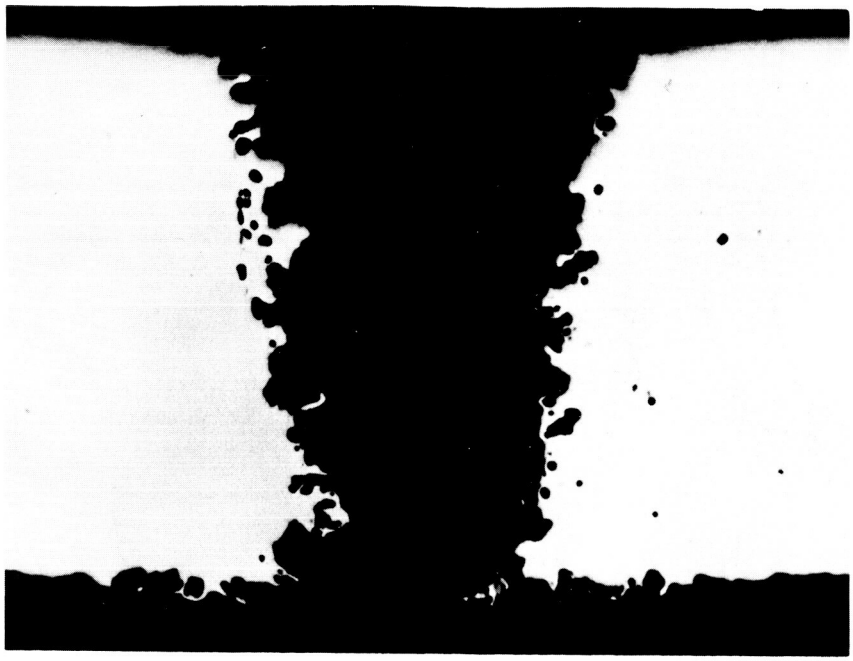
Microprobe analysis of the area near the braze zone of capsule No. 4 (1800°C - 1000 hours) showed that molybdenum and rhenium had diffused into the cladding from the braze zone. This result supports the hypothesis that porosity, which developed in the braze zone during isothermal testing, was caused by the Kirkendall effect.[5]

The microprobe analysis on capsules No. 7 (1800°C - 5000 hours) and 19 (2000°C - 3000 hours) revealed no uranium, aluminum, or cesium in the cladding and no aluminum, rhenium, tungsten, or cesium in the UO_2.

Of considerable interest in both capsules No. 7 and 10 were the metallic deposits found in the vent hole of each (Figures 20 and 21 respectively). A thin bridge of UO_2 or UO_2 and tungsten/rhenium may have existed across the narrow end of the vent hole of capsule No. 7. The reason for believing a bridge existed is that a number of loose particles of UO_2 containing metallic inclusions of tungsten/rhenium were formed in the mounting epoxy directly opposite and below the vent hole. The UO_2 may have been broken away from the bottom of the vent hole during metallographic mounting or sectioning procedures. No evidence of metallic inclusions of tungsten/rhenium were found anywhere else in the UO_2 inside the capsule.

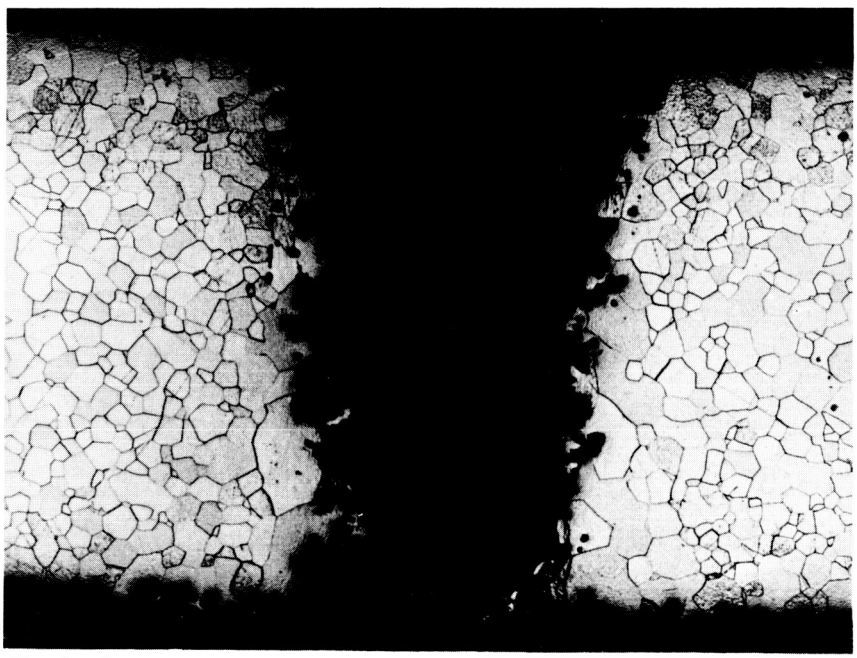
A thin metallic layer was found over the vent of capsule No. 10 (Figure 21). The same kind of cap was found on capsule No. 9. The layer of capsule No. 9 was not leaktight when tested with a helium mass spectrometer leak detector. The layer of both capsules 9 and 10 appeared continuous when observed before sectioning. The discontinuous appearance of the layer in Figure 21 was probably caused by the cutoff wheel during sectioning.

The vent deposits of capsules 7 and 10 contained nonmetallic inclusions which were identified as UO_2. A spectral scan of the nonmetallic phase also indicated the presence of a few tenths of a percent tungsten, but no rhenium.



As-Polished 100X

a.



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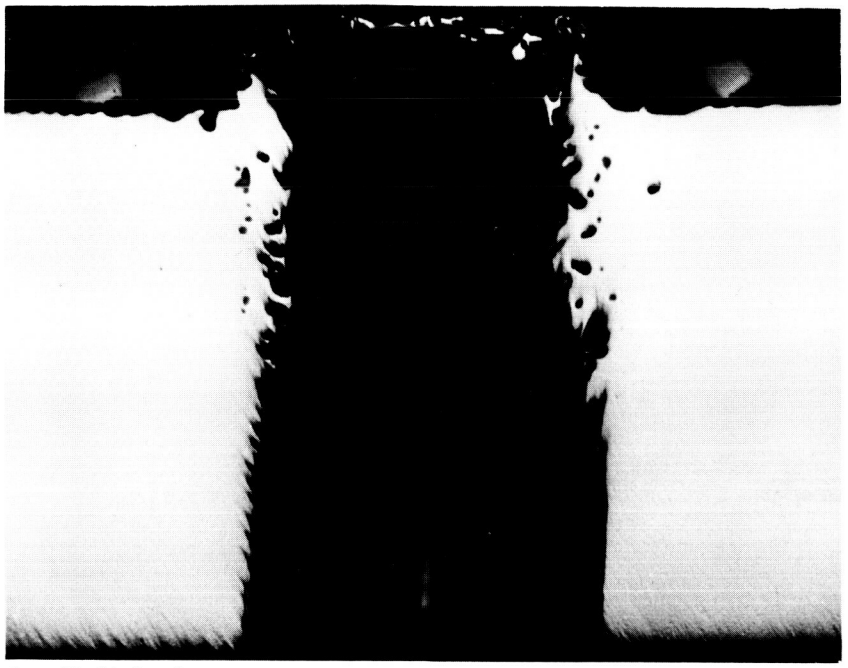
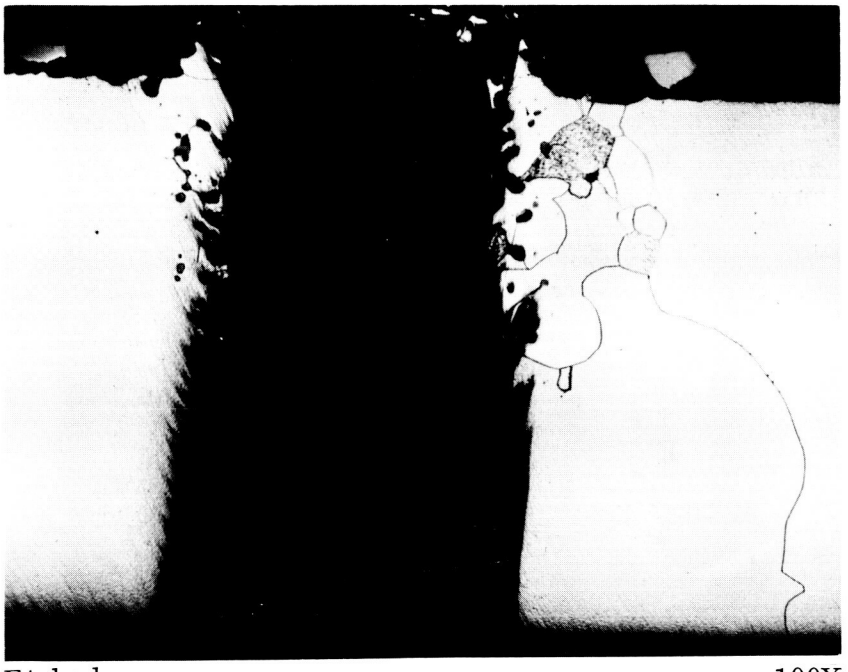
b.

FIGURE 20. VENT OF CAPSULE NO. 7. 1800°C - 5000 HOURS. THE URANIUM DIOXIDE PELLET (not shown) IS ADJACENT TO THE MINIMUM VENT DIAMETER



As-Polished 100X

a.



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b.

FIGURE 21. VENT OF CAPSULE NO. 10. 2000°C - 3000 HOURS. THE URANIUM OXIDE PELLET (not shown) IS ADJACENT TO THE MAXIMUM VENT DIAMETER

It was not possible to determine if the tungsten was in solution in the UO_2, or if the particle was so thin that the tungsten beneath the particle was excited. Spectral scans of the metallic deposits in both vents showed only tungsten and rhenium were present. Scans and spot checks of the clad caps, both in and near the metallic deposit of the vents, showed a continuously increasing gradient in the rhenium content of the cap extending perpendicularly from the edge of the metallic deposit in the vent to a distance of approximately 400 microns inside the cap. A scan of the rhenium content of capsule 10 is shown in Figure 22.

In capsule No. 7 the rhenium content varied from approximately 5 to 25% rhenium going from approximately 30 to 400 microns from the edge of the vent deposit. In capsule No. 10, the rhenium variation was from approximately 18 to 25% going from 10 to 400 microns distance respectively.

Because of the appearance of the vents of capsules No. 7 and 10, the other capsules previously examined were reground and their vents examined. For the 1800°C capsules the vents tend to show increasing metallic deposits with increasing time of test, except for the vent of capsule No. 2 (1800°C - 2000 hours) which showed no trace of buildup. This latter vent and the vent of capsule 1 was made by drilling rod stock from Hoskins. All other caps were supplied by Chase Brass, with vents made by electrodischarge machining. The vent of capsule No. 10 shows a buildup near the end of the vent farthest from the UO_2 pellet, whereas all the 1800°C capsules tended to have buildups on the side of the vent closest to the UO_2. The vent of capsule No. 12 (2200°C - 1000 hours) shows only a slight buildup of metallic phase and no visible UO_2.

The gradual constriction of the vents and the transport of UO_2 into the vent region was not fully understood.

The constriction did not develop to the point of sealing the vents for any of the capsules tested. The vents in all capsules were open to vision except

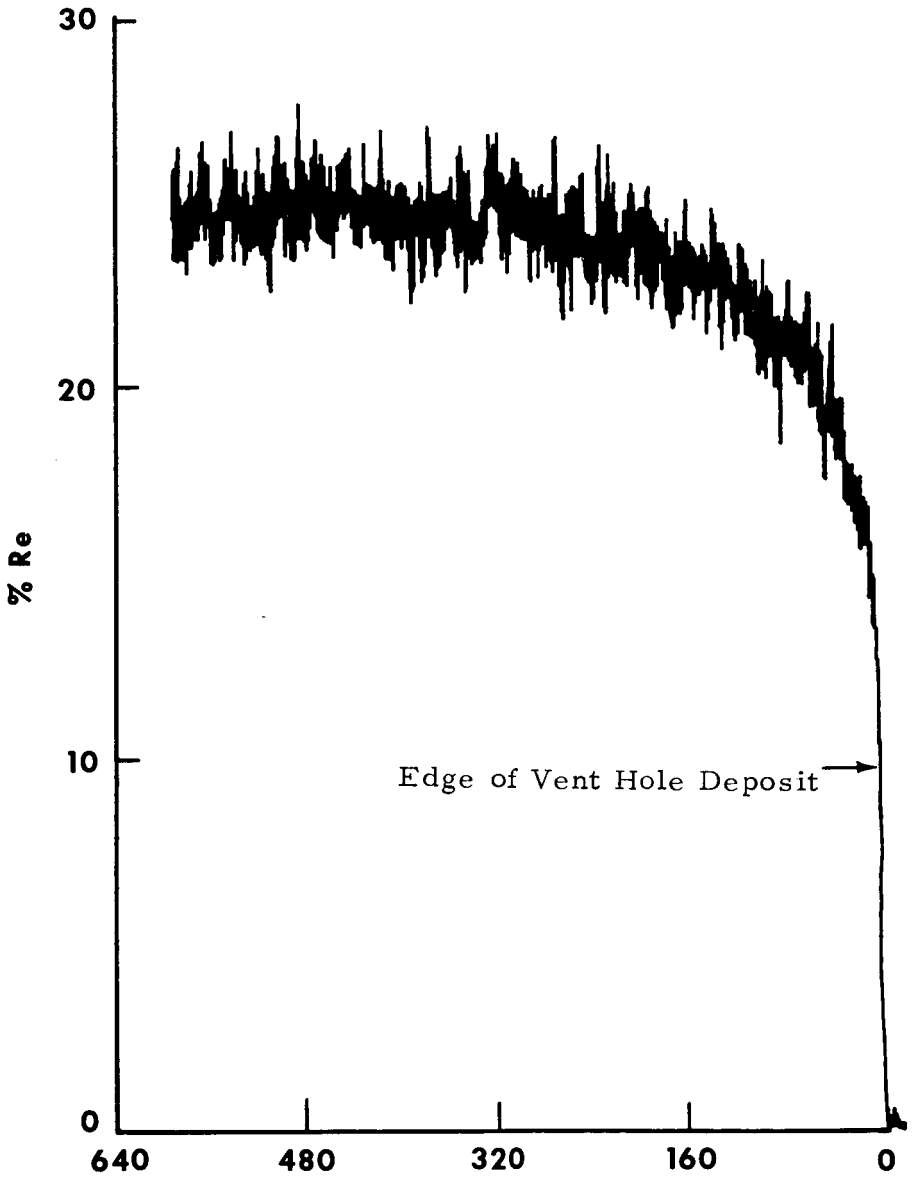


FIGURE 22. ELECTRON MICROPROBE SCAN OF RHENIUM IN TUNGSTEN/RHENIUM CAP OF CAPSULE NO. 10. (2000°C - 3000 hours)

capsule No. 10, and it passed helium freely.

In order to determine if the chemistry of the material had changed during testing, chemical analysis of the caps of capsules No. 10 (2000°C - 3000 hours) and 12 (2200°C - 1000 hours) and of as-received caps was performed and the results are reported in Table VI. The composition of the cladding has not changed much in either capsule except for a pick up in uranium content, a small pick up of oxygen and a considerable pick up of tantalum in capsule No. 12. The tantalum may have come form the tantalum chamber in which the capsules were housed during the test.

B. Thermal Cycling of Vented and Unvented Capsule in Vacuum Environment

Objective

The thermal-cycling experiments with UO_2 in W-25 wt % Re emitter containers were made to find if an undesirable amount of interaction between the fuel and emitter would occur with extensive thermal cycling. The concern was that the substoichiometry (UO_{2-x}) introduced at high temperature by oxygen diffusion, and the disproportionation of the substoichiometric phase to UO_2 and uranium at the lower temperature, would in some way lead to enhanced interaction. The free uranium released at the lower temperature might react with the W-25 wt % Re, and the remaining UO_2 could again lead to undesirable interactions.

Experimental Methods

Capsule Preparation - Capsules were prepared by using the same materials and the same fabrication methods described in Part I-A. The weight and dimensions of each sample were recorded before testing.

TABLE VI. CHEMICAL ANALYSIS TUNGSTEN-25% RHENIUM END CAPS

Element	As-Received	Capsule No. 10 (2000°C-3000 hours)	Capsule No. 12 (2200°C-1000 hours)	Analytical Technique
Re	24.89%	25.02%	25.08%	Gravimetric precipitation
O	40	140 ppm	95 ppm	Inert gas fusion
C	26	n.d.*	n.d.	Conductometric
N	100	n.d.	n.d.	Micro/Kjeldahl
Al	< 10	30	< 10	Emission spectrographic
Ca	< 1	4	3	Emission spectrographic
Cr	< 10	< 10	< 10	Emission spectrographic
Cd	< 20	< 10	< 10	Emission spectrographic
Cb	< 50	< 50	< 50	Emission spectrographic
Cu	20	27	26	Emission spectrographic
Cs	< 50	< 50	< 50	Emission spectrographic
Fe	7	25	17	Emission spectrographic
Hf	<100	<100	<100	Emission spectrographic
Mo	115	85	86	Emission spectrographic
Ni	5	7	5	Emission spectrographic
Na	< 5	15	15	Emission spectrographic
Si	< 20	25	< 20	Emission spectrographic
Sn	< 20	< 20	< 20	Emission spectrographic
Ta	<100	<100	1000	Emission spectrographic
Ti	1	2	1	Emission spectrographic
Zr	1	< 1	< 1	Emission spectrographic
Zn	<100	< 50	< 50	Emission spectrographic
U	< 0.5	360	48.2	Mass spectrometric
U (wall)	< 0.5	4.5	48.2	Mass spectrometric

*n.d. = no determination

Testing Procedures - Thermal-cycling tests were performed in an induction-heated (5kW, 400 kc) vacuum chamber operating with a vacuum between 10^{-5} and 10^{-6} torr. To reach desired temperatures, each capsule was enclosed in a cylindrical tungsten-tantalum susceptor concentrically supported within a copper water-cooled current concentrator. Temperatures were measured with an optical pyrometer.

Three different cycling modes were investigated and are illustrated in Figure 23. Approximately 1 minute of the first 10 minutes of cycles I and II was required to reach the maximum temperature from 725°C. One minute each was required for the heating and the cooling steps of cycle III. All cooling was by radiation from the capsule after the power was shut off.

Test Program - The types of thermal cycling tests are given in Table VII.

TABLE VII. THERMAL CYCLING TESTS OF UO_2 CLAD WITH W-25 WT % Re

Capsule	Vented	Type of Cycle	Period, min.	No. of Cycles	Post-Test Examination
A	Yes	I	15	50	Yes
B	No	I	15	50	No
C	Yes	II	15	50	Yes
D	No	II	15	50	No
E	Yes	III	10	50	Yes
F	No	III	10	50	No

Post-Test Evaluation - After thermal cycling, each capsule was measured and inspected. The three vented capsules were then examined metallographically, and Capsule E was studied with the electron microprobe.

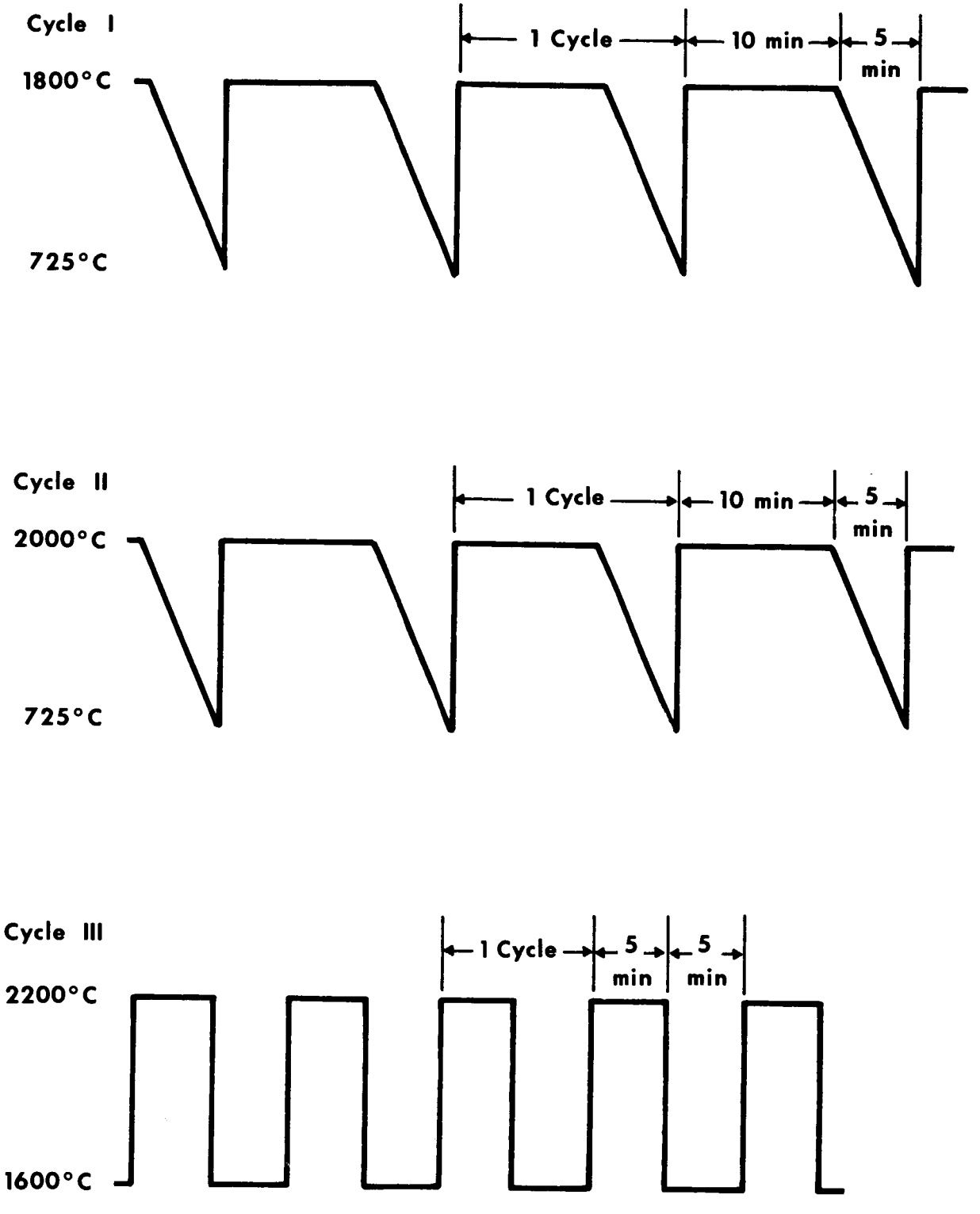


FIGURE 23. SCHEMATIC OF THERMAL CYCLING TEST PERFORMED IN VACUUM ON CAPSULES OF UO_2 CLAD WITH W-25% Re

Results and Discussion

Inspection and Measurements - No surface defects were found on any of the capsules after being inspected with Zyglo. The unvented capsules proved to be helium leaktight.

Values for the dimensional and weight changes of each capsule produced during thermal treatment are presented in Table VIII. The positive changes in outside diameter (all < 1%) are probably caused by the expansion of the UO_2 pellet. No unexpected changes occurred in the length of any capsule except for an unexplained decrease of 1.5% in the length of capsule E. Weight changes varied from a gain of 0.13% for capsule F to a loss of 0.82% for capsule E. The weight gain observed in some capsules may be attributed to a pickup of Ta. The accumulation of metallic deposits prevented accurate determinations of the weight of UO_2 lost from the vented capsules.

TABLE VIII. DIMENSIONAL AND WEIGHT CHANGES OF THERMALLY CYCLED SAMPLES

Capsule	Outside Diameter Change, mils	Length Change, mils	Weight Change, mg
A	+ 1	0	- 0.5
B	+ 2	0	+ 1.8
C	+ 2	0	+ 2.0
D	+ 2	+ 2	+ 14.4
E	+ 3	- 7	- 104.0
F	+ 1	+ 6	+ 15.7

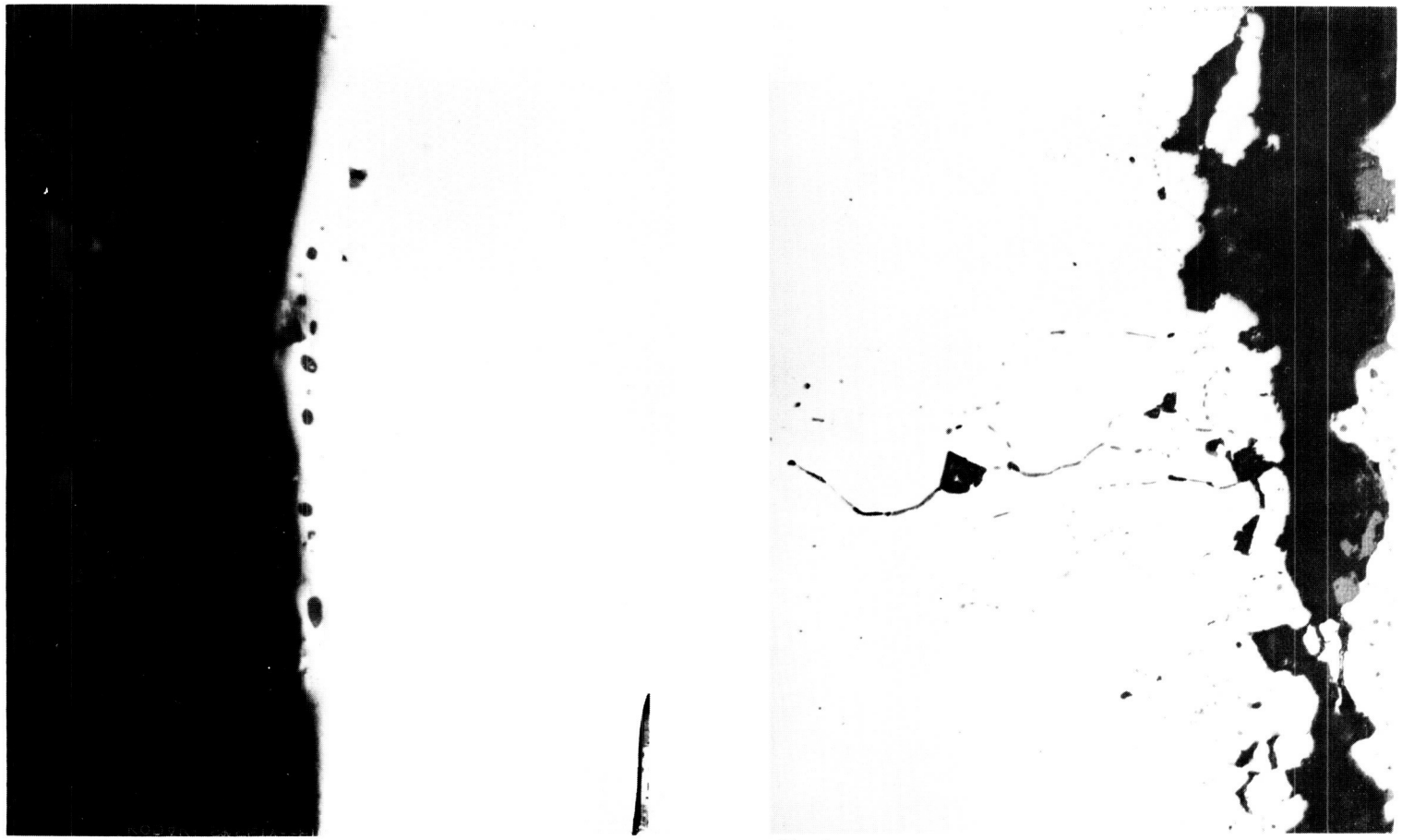
Metallography - All three vented capsules exhibited equiaxed W-Re grains free of sigma phase at 100X. The UO_2 grains were equiaxed in capsule A and C except for some vapor-deposited columnar grains in Capsule C. The columnar grains were caused by a small thermal gradient across the capsule during testing. The UO_2 vaporized from free surfaces (e.g., cracks) moved toward the cooler end of the capsule and deposited.

Capsule E was in a thermal gradient of 50°C from one end of the capsule to the other, and considerable redistribution of the UO_2 occurred by vapor transport. The gradient caused a large percentage of columnar grains and a large UO_2 weight loss.

A metallic phase (identified as uranium by microprobe analysis) was found in all three UO_2 samples. In capsules A and C most of the uranium was randomly distributed throughout the UO_2 as small particles within the grains. In capsule E, the uranium was located almost exclusively at UO_2 grain boundaries and pores.

No gross interaction between the UO_2 and the W-Re was observed in any of the capsules; however, traces of interaction were found in capsule E. Two isolated areas showed nonmetallic inclusions in the container (identified as UO_2 by microprobe analysis). One area near the edge of the container wall had inclusions up to 5 microns (Figure 24a). The other area near one weld gap had somewhat larger inclusions (Figure 24b). It is hypothesized that the UO_2 in the container wall was the product of an excess oxygen reaction. Small pits appeared at four or five locations along the container cap after etching. It was thought that these pits may have been areas where a second metallic phase had formed. Upon repolishing capsule E in preparation for microprobe analysis, these areas disappeared and could not be identified with the microprobe.

Microprobe Analysis - In addition to identifying the inclusions, an electron microprobe was used to detect interdiffusion of the container and fuel elements. Slow scan and spot checks of the container and UO_2 revealed no interdiffusion.



a. UO_2 (grey inclusions) found in clad wall

b. UO_2 (appears grey) found in clad above one weld gap

FIGURE 24. SAMPLE E CYCLED 2200-1600°C. TRACES OF INTERACTION

PART II. VACUUM ENVIRONMENTAL TESTING OF W-UO_2 CERMETS CLAD WITH W-25 WT % Re

The objective of Part II of this investigation was to determine the chemical compatibility of W-60 and 70 vol % UO_2 cermet fuels clad with W-25 wt % Re at 1800 and 2000°C for times to 1000 hours in a vacuum environment.

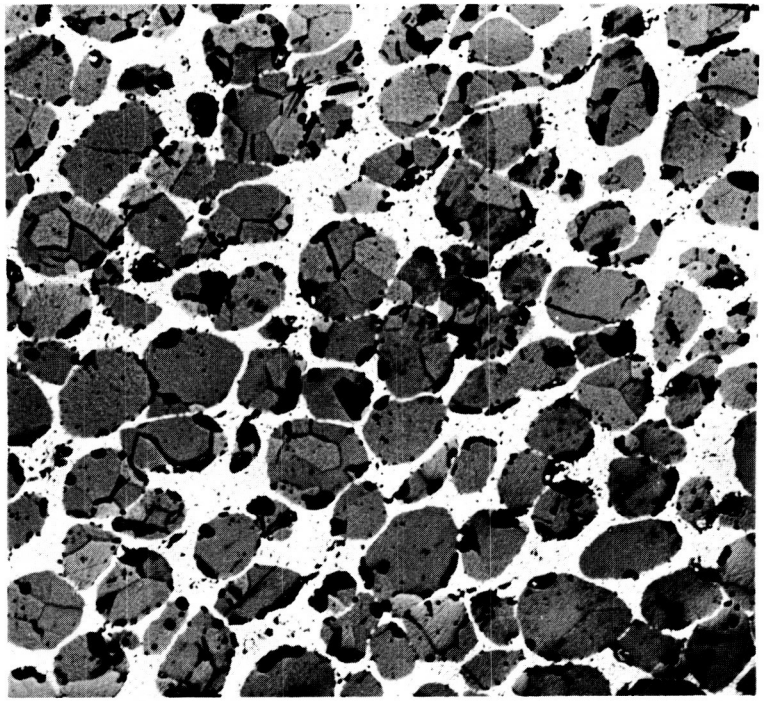
A. Experimental Methods

Capsule Preparation

Capsule Design - Test capsules* were cylindrical cermet cores of 60 or 70 vol % UO_2 in a continuous tungsten matrix clad with W-25 wt % Re alloy. The cermet cores were bonded to the clad and sealed in a high-temperature, pressurized helium autoclave. The clad capsules were approximately 0.49 inch long and 0.45 inch outside diameter.

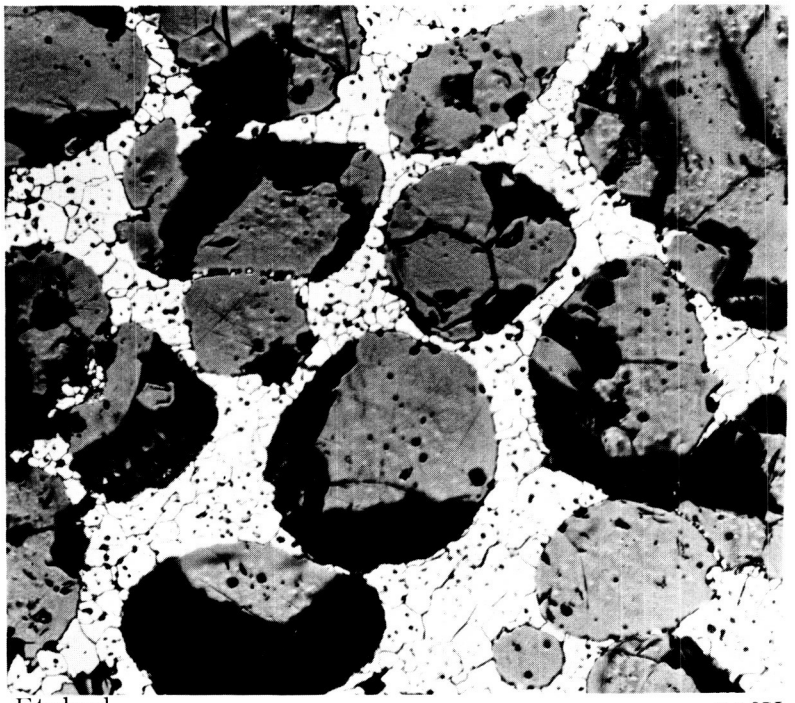
Fuel Fabrication and Characterization - Cermet cores of 60 and 70 vol % UO_2 in W were fabricated by blending and sintering powders of UO_2 and W. Chemical analyses of the tungsten and depleted UO_2 powders used are given in Table IX. The 60 vol % core was sintered at 2400°C for 1 hour in hydrogen, cooled to 1800°C and held for 18 hours, and furnace cooled to room temperature in helium. The extended treatment at 1800°C was given to promote densification; final core density was 93% of theoretical. The 70 vol % core was sintered at 2500°C for 1 hour in hydrogen, cooled to room temperature in a helium atmosphere in 2 to 3 hours, and had a final density of 89.7% of theoretical. The dimensions and weights of the cores used in each sample are given in Table IX. Micrographs of a typical core are shown in Figure 25.

*All test capsules were fabricated at General Electric Co., Nuclear Materials and Propulsion Operation, Cincinnati, Ohio.



As-Polished 100X

a.



Etched 250X

b.

FIGURE 25. TYPICAL W-60 vol % UO_2 CERMET FUEL. THE ROUGHLY SPHERICAL UO_2 PARTICLES (appears grey) ARE IMBEDDED IN A TUNGSTEN MATRIX (appears white). THE BLACK AREA IN SOME UO_2 PARTICLES IS BELIEVED TO BE PULLOUT

Clad Fabrication and Characterization - The W-25 wt % Re tube and rod used in fabricating the cermet fuel containers was structurally and chemically identical to the Hoskins W-25 wt % Re used for the UO_2-W-Re compatibility tests. The dimensions and results of inspection of the container are presented in Table X.

TABLE IX. CHEMICAL ANALYSIS OF W AND UO_2 POWDERS USED IN PREPARATION OF CERMET FUELS

Tungsten Powder

C	11 ppm (Leco analysis)	Na	26 ppm (flame photometry)
O	1063 ppm (Leco analysis)	K	69 ppm (flame photometry)

Tungsten Powder

Spectrographic Analysis (ppm)

Al	< 6	Cr	10
Ca	22	Ni	6
Si	11	Cu	6
Mo	60	Mn	< 6
Fe	170	Mg	5
		Sn	< 6

UO_2 Powder

Spectrographic Analysis (ppm)

Al	< 25	Fe	<150
B	< 1	Mn	< 10
Cd	< 1	Ni	<150
Cr	<100	Pb	< 10
Cu	< 10	Si	< 50
F	150	Ag	< 0.1
		Sn	< 2

Capsule Assembly - Cermet cores were inserted inside the W-Re tube, end caps were put in place, and the assembly isostatically gas-pressure bonded in an autoclave with approximately 10,000 psi helium at 1650°C. The capsules were heated to temperature 3 hours, held at temperature for 2 hours and then cooled to room temperature overnight (~ 16 hours) under the 10,000 psi helium pressure.

As-Bonded Capsule Inspection and Measurements - After isostatic bonding, capsules were helium leak checked, Zyglo inspected, radiographed, ultrasonically tested, and inspected for evidence of alpha particle emission. The radiographic examination was made to determine the uniformity of density and to detect any internal cracks in each capsule. Transverse internal cracks

TABLE X.[++] DIMENSIONS AND WEIGHTS OF CORE AND CLAD BEFORE AUTOCLAVING

		As-Sintered Core			W-25 wt % Re Cladding As-Received		
Capsule	UO_2 vol%	Core Weight, gm	Core Length, inch	Core Diameter, inch	Tube Wall, inch	Tube o.d., inch	End Plugs Thick, inch
VWR-1 *	60	8.5231	0.3000	0.4061	0.021	0.447	0.100
VWR-2 *	60	8.5357	0.3000	0.4060	0.021	0.447	0.100
VWR-3 *	60	8.4878	0.2995	0.4055 0.4062	0.21	0.447	0.100
VWR-4 * +	60	10.1731	0.3600	0.4060 0.4065	0.021	0.447	0.060 ** 0.080
VWR-5 * +	70	11.6098	0.4098	0.4061 0.4062	0.021	0.447	0.040 0.050
VWR-6 *	60	8.6368	0.3003	0.4062	0.021	0.447	0.100
VWR-7 *	70	7.7340	0.300	0.4061 0.4063	0.021	0.447	0.100
VWR-8 *	70	7.7832	0.2998	0.4055 0.4060	0.021	0.447	0.100
VWR-9 *	70	7.6150	0.300	0.4055 0.4060	0.021	0.447	0.100
VWR-10 *	70	7.6393	0.2996	0.4055 0.4059	0.021	0.447	0.100

*Vacuum leak test OK; Zyglo test OK.

**Indicates thickness of each end cap when they were not identical.

+Zyglo test showed small pits.

++All measurements were performed by the supplier.

(probably in the core) were found in capsules VWR-2, -7, -9, and -10. A radiograph of the ten capsules revealed uniform density of each capsule. The ultrasonic test measured the degree of bond integrity of the core to the container. The results of each of the preceding inspections and the as-bonded capsule dimensions and weight are given in Table XI. An hourglass shape of the capsule indicated by the capsule dimensions was produced during autoclaving.

As-Bonded Capsule Metallography - Micrographs of the interfacial region of a typical as-bonded capsule are shown in Figure 26. Etching with Murakami's reagent revealed that the boundary between the core and container was irregular and contained an unidentified second phase. (Figure 26b). This second phase is clearly shown in Figure 27, where one area of the boundary between the core and container is shown in both the as-polished and etched conditions.

Testing Procedures - Capsules were isothermally treated in the same furnaces used for the UO_2-W-Re compatibility studies, but were supported outside the cesium chamber on a concentric tantalum stand. The environment of the cermet capsules during testing was a vacuum of 10^{-6} to 10^{-7} torr.

Test Program - Ten tests were planned as shown in Table XII. All capsules were unvented.

TABLE XII. TEST PROGRAM PLANNED FOR W-UO_2 CERMETS CLAD WITH W-25 WT % Re (10^{-6} torr vacuum)

Capsule	UO_2, vol %	Temperature, °C	Test Time, hours
VWR-3	60	1800	500
VWR-5	70	1800	500
VWR-4	60	1800	1000
VWR-8	70	1800	1000
VWR-6	60	1800	>1000
VWR-2	60	2000	500
VWR-9	70	2000	500
VWR-1	60	2000	1000
VWR-7	70	2000	1000
VWR-10	70	2000	>1000

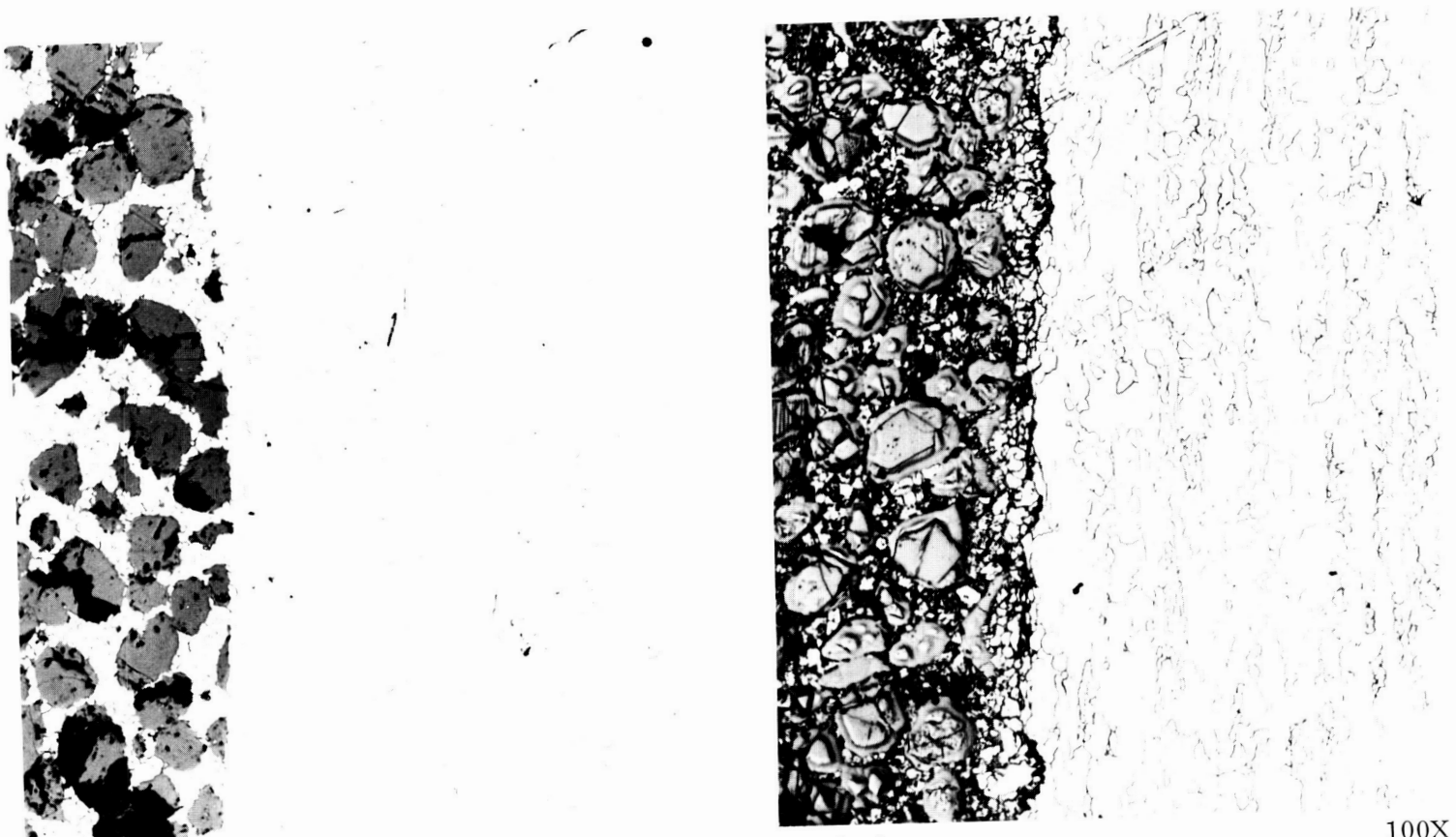
TABLE XI.[++] AS-BONDED CAPSULE INSPECTION, DIMENSIONS AND WEIGHT

Capsule	Final Dimensions Length,	Diameter, inch Top,	Center,	Bottom,	Specimen Weight After Autoclave, gm
VWR-1 *	0.4964	0.4483	0.4357	0.4488	21.3273
VWR-2 * +	0.4966	0.4483	0.4373	0.4483	21.3552
VWR-3 *	0.4963	0.4483	0.4368	0.4485	21.3288
VWR-4 *	0.4948	0.4485	0.4373	0.4483	20.4564
VWR-5 *	0.4940	0.4480	0.4357	0.4484	19.8080
VWR-6 *	0.4977	0.4473	0.4359	0.4485	22.3673
VWR-7 * +	0.4956	0.4475	0.4307	0.4468	20.5068
VWR-8 *	0.4943	0.4468	0.4297	0.4475	20.4337
VWR-9 * +	0.4930	0.4473	0.4289	0.4478	20.3343
VWR-10 * +	0.4923	0.4475	0.4287	0.4475	20.3788

*Helium leak check, 300 psi, OK; Bond Check, OK; Alpha Check, OK; X-ray, OK; Zyglo Test, OK, except for VWR-1 - small pit.

*+X-ray showed core cracked.

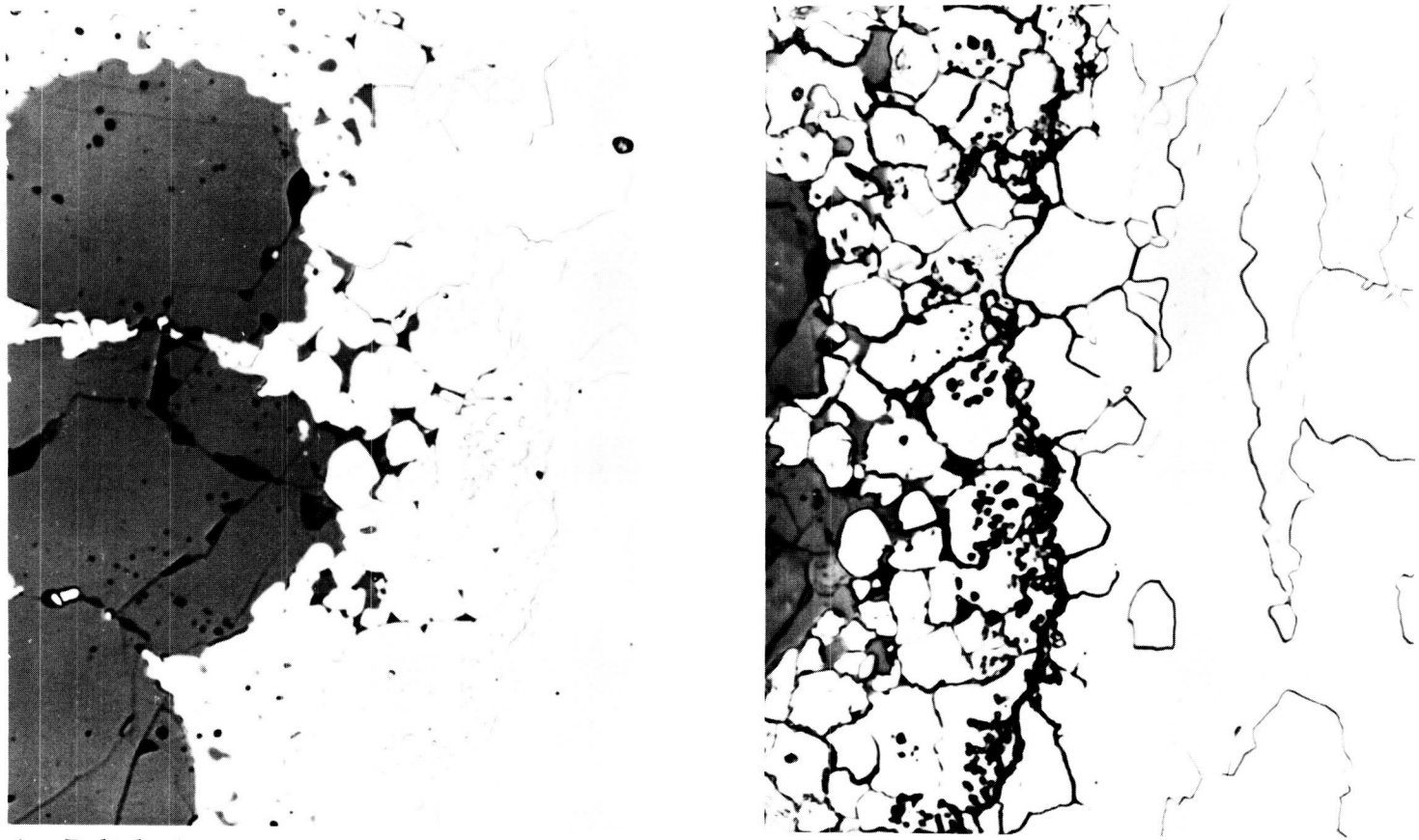
++All measurements and inspection procedures were performed by the supplier.



As-Polished 100X Etched 100X

a. b.

FIGURE 26. TYPICAL W-60 vol % UO_2 INTERFACIAL AREA BETWEEN AS-BONDED CERMET CORE (left) AND W/Re CLAD (right)



As-Polished 500X

a.

500X

b. Same area as (a) etched 20 sec. with Murakami's reagent

FIGURE 27. W-60 vol % UO_2 - EVIDENCE OF SECOND PHASE IN THE CORE/CLAD CORE (left) AND W/Re CLAD (right) BOUNDARY OF AS-BONDED CAPSULE

Post-Test Evaluation - Visual inspection and weight and dimensional measurements were performed for each capsule after thermal testing. Microprobe analysis and metallography were done in a manner similar to that discussed previously in this report.

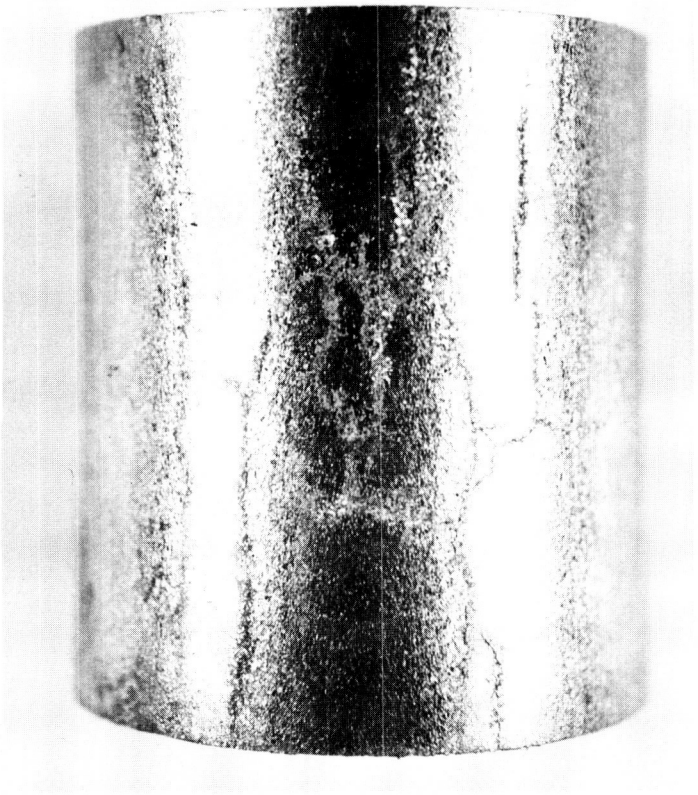
B. Results and Discussion

Tests were terminated when the shutdown to examine the capsules tested for 500 hours at 1800°C showed extensive container cracking in the capsule. Immediate inspection of the other test capsules, which had been exposed to only 389 hours at 2000°C, revealed similar container failures in all the capsules.

Inspection and Measurements - Longitudinal and circumferential cracks in tube walls and radial cracks in the caps were found in all capsules. Cracking was most severe in capsules tested at 2000°C. Macrophotographs of a typical capsule tested at 1800°C are shown in Figure 28. The bright areas around the cracks have been analyzed by x-ray diffraction and x-ray fluorescence. Analysis indicated tungsten, rhenium, and traces of UO_2 and tantalum. The tantalum was from a tantalum stand on which the capsules rested during furnace treatment.

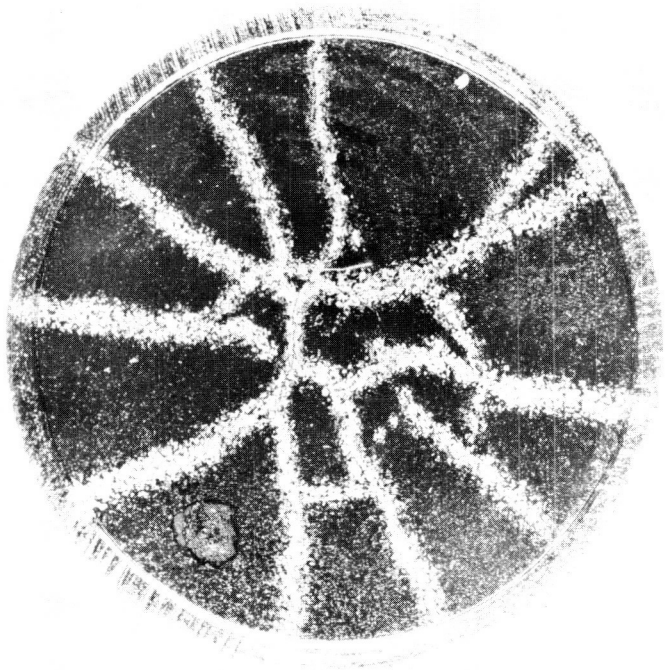
The weight and dimensional changes of each capsule caused by thermal testing are presented in Table XIII. The weight changes vary from a low of $\sim$ + 0.2% (VWR-4) to a high of $\sim$ - 7.5% (VWR-1). The considerable variation among the capsules was attributed to differences in temperature, extent of cracking, and pickup of tantalum from the support stand. The large loss of weight from VWR-6 compared to other capsules tested at 1800°C, may have resulted from an error in the pre-test weight measurement.

All dimensions of a given capsule increased; the greatest increases occurred for the capsule tested at 2000°C. The length increased from a low of $\sim$ 0.4% (VWR-5) to a high of $\sim$ 2.4% (VWR-10). The central outside



7X

a. Side view showing longitudinal and circumferential cracks



7X

b. View of radial cracks in top cap of capsule

FIGURE 28. MACROPHOTOGRAPHS OF TYPICAL CERMET CLAD CAPSULE TREATED AT 1800°C FOR 500 HOURS

diameter varied from a low of ~ 0.6% (VWR-4) to a high of ~ 2.4% (VWR-1).

TABLE XIII. WEIGHT AND DIMENSIONAL CHANGE OF CAPSULES

Capsule	Weight Change, gram	Length Change, mils	Outside Diameter Change, mils		
			Top	Center	Bottom
VWR-3 *	+ 0.0106	+ 4.7	+ 0.7	+ 3.2	+ 0.5
VWR-5 *	- 0.0003	+ 2.0	+ 1.7	+ 4.3	+ 2.6
VWR-4 *	+ 0.0410	+ 5.2	+ 0.5	+ 2.7	+ 1.7
VWR-8 *	+ 0.0129	+ 3.7	+ 0.2	+ 4.3	+ 0.5
VWR-6 *	- 1.0057	+ 4.3	+ 0.7	+ 3.1	+ 1.5
VWR-2 **	- 1.0806	+ 8.4	+ 1.7	+ 3.7	+ 4.7
VWR-9 **	- 0.6929	+ 9.0	+ 3.7	+ 8.1	+ 5.2
VWR-1 **	- 1.6249	+ 6.6	+ 2.7	+10.3	+ 2.2
VWR-7 **	- 0.4260	+ 10.4	+ 3.5	+ 7.3	+ 4.2
VWR-10 **	- 0.7740	+ 11.7	+ 4.5	+ 8.3	+ 4.5

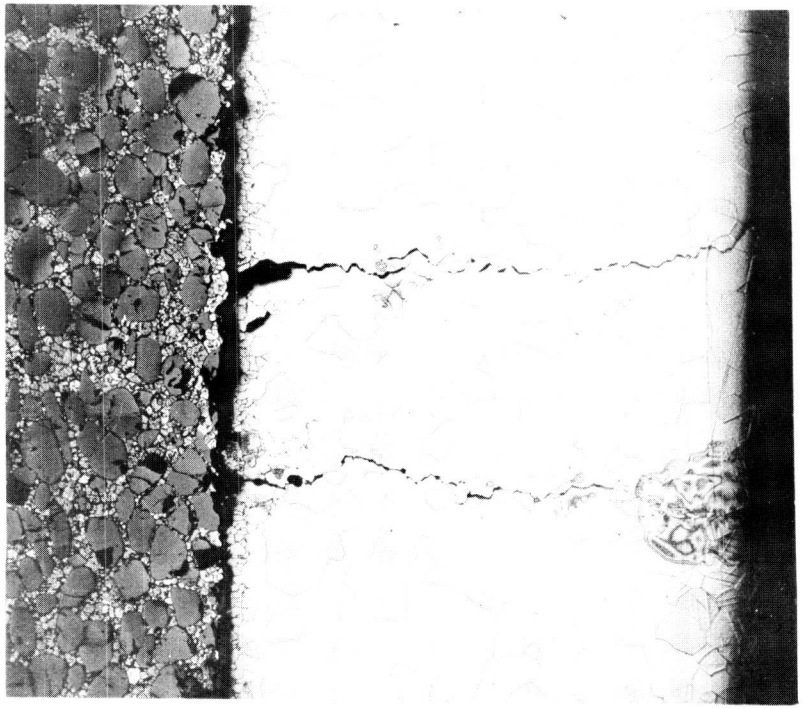
*Test Time - 500 hours at 1800°C

**Test Time - 389 hours at 2000°C

Metallography - One capsule of each UO_2 volume percent tested at 1800 and 2000°C was sectioned and examined metallographically.

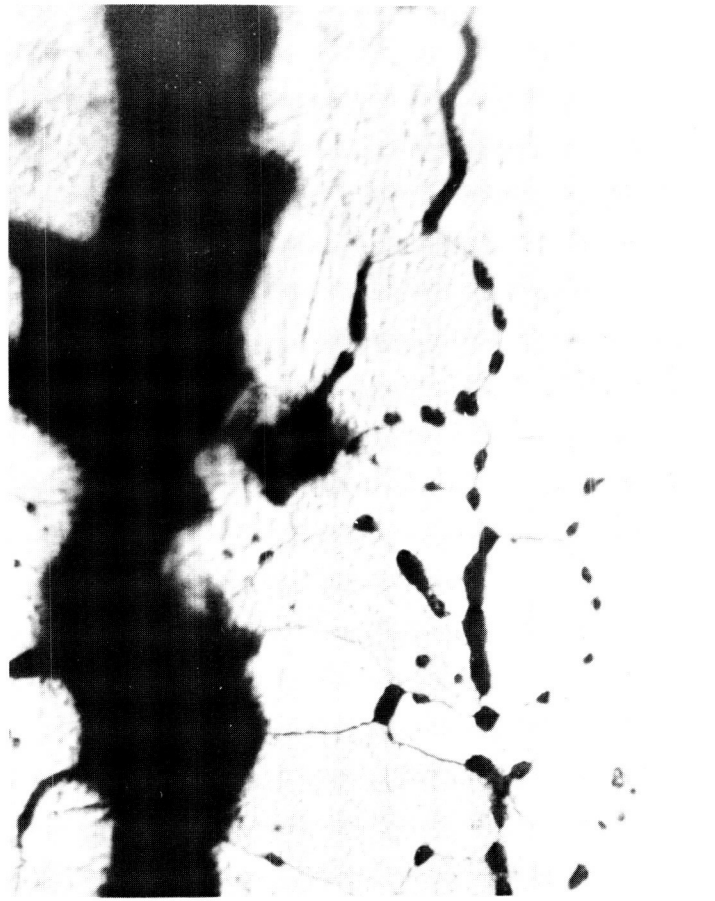
Figure 29 shows interfacial areas of the core and the container of capsule VWR-5 (70 v/o UO_2) tested at 1800°C. A narrow zone of fine-grained metallic phase containing small intergranular nonmetallic inclusions (possibly UO_2) was found along the interfacial edge of the container. This zone extended completely around the inner edge of the container and is more clearly shown at 1000X in Figure 29b. The core structure revealed UO_2 penetration into the tungsten grain boundaries.

Hardness measurements were made on the container, the grains of the zone, and the tungsten core matrix. The zone proved to be intermediate in hardness (396 DPH) between that of the container (438 DPH) and the tungsten core matrix (360 DPH). In VWR-3 nonmetallic phase (possibly UO_2) was observed in the grain boundaries in the W-25 w/o clad.



Etched 50X

a.



Etched 1000X

b.

FIGURE 29. INTERFACIAL AREA OF CERMET/CLAD CAPSULE VWR-5 ANNEALED AT 1800°C FOR 500 HOURS. THE W/Re CLAD APPEARS WHITE

The black area between the core and the container was void and like the unidentified zone mentioned before, was continuous around the internal periphery of the container. The void was produced during thermal treatment and may have resulted from: (1) the core breaking away from the container; (2) the evaporation of UO_2 from this region through cracks leading to the outside surface of the cladding; or (3) both these mechanisms. It is most likely that both mechanisms were operative. Interpretation (2) required that the UO_2 be continuously connected. Examination of the microstructure of the rough area around the periphery of the core clearly shows that the UO_2 has been interconnected because it escaped the capsule and left behind a skeleton of tungsten.

The reason for the capsule failures is not definitely known, but post-test observations strongly indicate the main cause was the large difference in the amount of expansion between the core and the container, which gave rise to high triaxial stresses in the sample container. Data [8] indicate that a 60 vol % UO_2 core exceeds the linear thermal expansion of W-25 wt % Re container at 1800 and 2000°C by ~ 30%. The capsules tested at 1800 and 2000°C received 2 and 15 inadvertent thermal cycles, respectively. (Drops in the H_2O line pressure caused automatic safety shutdowns). The more extensive cracking of the 2000°C capsule compared to the 1800°C capsule was probably related to the larger number of cycles the former received. The additional cycles permitted greater opportunity for UO_2 to move into container grain boundaries and pre-existing cracks. Further cracking occurred by a wedging action produced by the larger volume expansion of UO_2 and uranium compared to that of the container.

Microprobe Analysis - Microporbe analysis was performed on capsule VWR-5 to identify the extent of interdiffusion between the core and the clad, metallic phases of the interfacial zone, and the composition of the container near surface cracks. No free uranium was found in the W-25 wt % Re container, in the metallic grains of the interfacial zone, or in the tungsten grains of the core matrix. Rhenium content varied from 24 to 26% in the container

except within 100 microns of the interface between the container body and the zone. From this distance to within the metallic grains of the zone the Re content decreased from 26% to 11%. Tungsten was the only other element detected in the grains of the zone. No Re was found in the tungsten grains of the core matrix. Tantalum was found at the outer edge of the W-25 wt % Re. The concentration varied from 3 to 10% at the outer surface to a non-detectable amount 25 microns into the container from the surface. Tantalum ($\sim$ 5%) was also found near a crack in the area where the crack intersected the outside surface of the cladding. Tungsten and 22-24% Re were the only other elements detected in this area. No tantalum was found in the main body of the container, in the metallic grains of the zone, or in the tungsten grains of the core matrix. The nonmetallic particles observed in the grain boundaries of the zone and also penetrating some grain boundaries of the container were UO_2. The metallic particles in the UO_2 of the core which were large enough to microprobe were identified as tungsten.

The Re concentration gradient in the W-25 wt % Re near the zone and the low Re content of the zone grains indicate that Re diffused into the zone from the container. This observation plus the similarity in appearance of the structure of the zone and the core, strongly suggest that the zone was originally part of the core. The void area, described in the metallography section, was thus caused by a separation of the core from itself by the mechanisms discussed.

The tantalum found near the external surface of the cladding was undoubtedly caused by some evaporation from the tantalum stand on which the capsules were supported. The absence of tantalum in the interior of the capsule indicates that tantalum did not contribute to the failures of the capsule.

The microprobe results provide no evidence to contradict the belief that the capsule failures were probably caused by the thermal expansion mismatch of the core and the container.

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