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ORNL-4569

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**POSTIRRADIATION EXAMINATION AND
EVALUATION OF TUNGSTEN URANIA CERMETS**

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OAK RIDGE NATIONAL LABORATORY

operated by

UNION CARBIDE CORPORATION

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U.S. ATOMIC ENERGY COMMISSION

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Contract No. W-7405-eng-26

METALS AND CERAMICS DIVISION

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TUNGSTEN-URANIA CERMETS

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JUNE 1970

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Oak Ridge, Tennessee
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POSTIRRADIATION EXAMINATION AND EVALUATION OF
TUNGSTEN-URANIA CERMETS

A. E. Richt M. M. Martin
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ABSTRACT

Oak Ridge National Laboratory has completed the post-irradiation evaluation of tungsten-urania and tungsten-urania-thoria dispersions irradiated in the Oak Ridge Research Reactor by the Missile and Space Division of General Electric. Eight capsules were irradiated in the range of 1500 to 1730°C and with a burnup of approximately 1.4×10^{20} fissions/cm³ of the composite core. These capsules were irradiated to study the effect of type and magnitude of voids in reducing swelling of W-UO₂ and W-UO₂-ThO₂ cermet clad with a W-Re-Mo alloy.

All phases of the postirradiation examination program including burnup determination, dimensional measurements, and metallographic examination have been completed. The maximum increase in diameter of all eight pins ranged between 1.3 and 6.7%. Analysis of the irradiation data indicates void deployment to be less important than the total void content. End plenum void volume may have an upper effective limit of about 14%. Failure of one fuel capsule during irradiation may have been premature and caused by excessive mass transfer between fuel and cladding.

INTRODUCTION

In selecting a high-temperature fuel element for space power reactors, one must consider both bulk ceramic and the refractory metal cermet systems. In evaluating the advantages and disadvantages of either system, irradiation swelling for these fuels is always compared. Swelling is largely the result of the creation of fission gases, and the effectiveness of controlled void volume, void deployment, and venting are important fuel element design and fabrication criteria.

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Refractory metal cermets have been studied by several investigators.^{1,2} Generally the concept of a dispersion fuel is to contain fission products by surrounding each of the ceramic fuel particles with a high-strength metal. The metal matrix helps to restrain fuel swelling, lowers the fuel temperature by increasing the thermal conductivity of the composite fuel and providing thermal contact with the cladding, and decreases the temperature drop across the fueled body. Cermet fuels, however, have the disadvantage of lower specific uranium contents.

Internal porosity is used in both bulk fuel³ and plate-type cermets⁴ to reduce fuel swelling. If refractory metal cermets can use an end plenum in which to collect the gaseous fission products instead of an increased porosity within the cermet body, the difference between specific uranium contents of bulk ceramic and cermet fueled capsules is greatly reduced. However, for the end plenum to be effective, the cermet must have sufficient permeability. High-density cermet cores may not have sufficient permeability, and small vents may be necessary to facilitate movement of the fission gases into the end plenum. These vents further reduce the specific uranium content and would not be used unless they are absolutely necessary.

If end plenums, with or without vents, are effective in improving the dimensional stability of cermet fuels during irradiation, properly selected cermet fuel systems may be very competitive for space power reactors.

The purpose of this irradiation experiment was to study the effectiveness of a built-in end plenum within the range of 0 to 26 vol % for reducing swelling of W-Re-Mo clad UO_2 - and UO_2 - ThO_2 -bearing tungsten

¹D. L. Newsom and J. F. Collins, Irradiation Testing and Evaluation of Refractory Metal Fuel Elements of 710 Fast Reactor Program, GEMP-710 (October 1969).

²E. O. Speedel, D. E. Keger, and D. L. Keller, Preparation and Properties of UO_2 , UN, and UC Cermet Fuels, BMI-1842 (June 1968).

³Donald L. Keller and Walston Chubb, Progress on High-Temperature Fuels Technology During August Through October 1969, BMI-1875.

⁴Fuels and Materials Development Program Quart. Progr. Rept. Dec. 31, 1969, ORNL-4520.

cermet fuel specimens with 2 to 17% void volume or internal porosity within the core. The irradiations were conducted by the Missile and Space Division of General Electric in the Oak Ridge Research Reactor (ORR).

The postirradiation examination and evaluation were performed by the Metals and Ceramics Division at ORNL. The scope of the postirradiation examination program included burnup analysis, irradiation temperature history, dimensional measurements of the capsule, and metallography. All phases of this postirradiation program have been finished as described in this report.

DESCRIPTION OF TEST SPECIMENS

For the in-reactor experiment, eight clad fuel specimens were fabricated with void deployment in the fueled cores and built-in plenum regions as the principal variables. Figure 1 illustrates the longitudinal

Fig. 1. Longitudinal Cross Section of ORRF-5 Test Specimens Showing Integral Plenum Region. Compositions shown are volume percentages.

cross section of each of the specimens and gives their core compositions and total internal void contents in volume percent. The annular fuel core is about 1.19 cm OD $\times$ 0.45 cm ID $\times$ 3.3 cm long and is clad on both circumferential surfaces and ends with 0.038-cm-thick W-30 at. % Re-30 at. % Mo. This cladding composition is sometimes referred to as the 306 tungsten alloy. The overall length of the specimens varied from 5.08 to 6.65 cm depending on the length of the end plenum. With two exceptions, the void volume of the plenums was located at one end of the specimen as a single chamber. One high-density W-UO₂ specimen (336-5) with a total void content of 30% places one-third of the plenum void volume in the bore of the specimen. Another specimen (554-14) contains eight 1.1-mm-diam holes drilled longitudinally into the core to a depth of 2.54 cm; these holes account for 26% of the total 19 vol % internal void content.

The fabrication procedures for the test specimens have been reported by General Electric.⁵⁻⁸ In general, the essential processing steps are (1) preparation of a fuel agglomerate of -100 +230 mesh particle size from enriched UO₂, depleted UO₂, and (in six specimens) ThO₂, (2) blending of the fuel agglomerate with tungsten powder having an average particle size of 0.8 μ m, (3) pressing at 30,000 psi in a mechanical die, (4) sintering to achieve stoichiometric UO₂, (5) machining, (6) hot autoclaving for dimensional stability, (7) machining to size, (8) cleaning and vacuum outgassing, and (9) hot pressure bonding. Table 1 tabulates the detailed core composition, theoretical core density, and ²³⁵U content of each specimen.

⁵AEC Fuels and Materials Development Program, Eighth Annual Report, GEMP-1011, pp. 131-134 (Feb. 28, 1969). CLASSIFIED

⁶AEC Fuels and Materials Development Program, Seventh Annual Report, GEMP-1003, pp. 59-61 (Feb. 29, 1968). CLASSIFIED

⁷High-Temperature Materials Program, Sixth Annual Report, Part B, GEMP-475B, pp. 72-74 (Feb. 28, 1967). CLASSIFIED

⁸High-Temperature Materials Program, Fifth Annual Report, Part B, GEMP-400B, pp. 57-60 (Feb. 28, 1966). CLASSIFIED

Table 1. Cermet Composition, Theoretical Density, and ^{235}U Content of ORRF-5 Test Specimens

Composition	Specimen	Composition of Cermet, wt %		Composition of Fuel Agglomerate, wt %			Theoretical Core Density (g/cm ³)	Total ^{235}U Content (g)
		W Matrix	Fuel Agglomerate	UO ₂ Enriched ^a	UO ₂ Depleted ^b	ThO ₂		
336	{ 2 5	54.00	46.00	4.49	95.51		14.296	{ 0.673 0.668
554	{ 4 12 13 14	54.22	45.78	6.92	83.88	9.20	14.238	{ 0.954 0.958 0.944 0.897
836	{ 1 9	54.36	45.64	9.86	74.71	15.43	14.200	{ 1.223 1.212

^a70.05% ^{235}U in the uranium.

^b0.20% ^{235}U in the uranium.

Changes from their standard composition and fabrication procedure allowed General Electric to produce the three levels of void concentration of about 2, 9, and 16 vol % noted in Fig. 1. For example, the high-density specimens of the 336 composition were clad by standard pressure bonding of 700 kg/cm² at 1750°C for 3 hr. The fuel agglomerates of UO₂ for this composition were also fired at the standard temperature of 750°C. On the other hand, addition of ThO₂ to UO₂ and firing the resultant fuel agglomerate at 1600°C helped produce the lower density of the specimens from the 554 and 836 compositions. The sole purpose of the ThO₂ additions was to increase the void content of the cermet. The cores of intermediate density from the 554 composition (about 5 vol % ThO₂) were bonded at a reduced pressure of 350 kg/cm². Bonding at 1650°C and 315 kg/cm² (for the specimens from the 836 composition, which contained about 8 vol % ThO₂) produced the lowest core density.

IRRADIATION TEST CONDITIONS

The irradiation of GE's experiment ORRF-5 was conducted in the F-2 position of the ORR. Maximum unperturbed thermal neutron flux in this position is 2.2×10^{14} neutrons cm⁻² sec⁻¹. The cartridge assembly that contained the eight specimens during testing was basically the same as that for GE experiment ORRF-4 described elsewhere.⁹ The essential difference was in the design of the cartridges in that the specimens were located in pairs instead of being by themselves in the four stainless steel specimen tubes. The paired specimens were separated about 2 in. in the tubes, and the tubes were clustered. Water cooled the outside of the tubes while He-5% H₂ at about 20 cm³/min was used to transfer heat and protect the surface of the specimens. As required, neon was added to the gas mixture to maintain the desired test temperature.

Each specimen tube had its own gas supply and separate effluent gas line. These desirable features of the cartridge permitted individual fission gas monitoring and temperature regulation of any particular

⁹AEC Fuels and Materials Development Program, Seventh Annual Report, GEMP-1003, p. 85 (Feb. 29, 1968). CLASSIFIED

pair of specimens. The clustered assembly of tubes was also equipped with an insertion-retraction mechanism, which provides another means of temperature control by raising and lowering the entire assembly with respect to the horizontal midplane of the reactor core. Each specimen was instrumented with two thermocouples of W vs W-25% Re stranded wire insulated with BeO. As shown in Table 1, the fuel loadings were adjusted so that all eight specimens would operate in the range of 1500 to 1650°C. The thermocouples were located in the bore of each specimen, and all performed well throughout the duration of the test.

Irradiation testing of the specimens began February 18, 1969, and was terminated on July 31, 1969, when fission-product activity was detected in the coolant gas. The specimens operated for about 3400 hr at temperatures above 1250°C and achieved burnups in the range of 1.3 to 1.4×10^{20} fissions/cm³.

During the period of testing, the temperature of the specimens was recorded every morning Monday through Friday and when scheduled and unscheduled shutdowns occurred. Selected thermocouples were usually read when the position of the assembly was changed or when neon was added to the coolant gas. Figures 2 and 3 show the chronological temperature histograms for each specimen. In these plots, the vertical lines represent either a shutdown or a startup of the reactor. We note that the temperature of the specimens dropped below 800°C only 29 times during the test.

Figure 4 summarizes the temperature history shown in Figs. 2 and 3 for the actual operating time. The plots of the stated temperature against the accumulated times are quite useful in selecting and/or defining an average irradiation temperature and readily show the maximum irradiation temperature.

POSTIRRADIATION EXAMINATION

After about a month's decay, the test assembly was transferred to the hot cells for disassembly and examination. Nothing unusual was observed during disassembly. The specimen coolant tubes were free of obstruction and blockage, the specimens were easily removed from the

Fig. 2. Temperature Histogram for GE-NSP Tungsten-UO₂ Cermet Irradiations.

Fig. 3. Temperature Histograms for GE-NSP Tungsten-UO₂ Cermet Irradiations.

Fig. 4. Temperature Distribution of ORRF-5 Test Specimens, Showing Accumulated Time Above Stated Temperature. Experiment operated above 800°C for a total duration of 3436 hr.

holders, and all 16 specimen thermocouples appeared to be in excellent physical condition.

The overall appearance of the test specimens after irradiation is shown in Fig. 5. In general, all specimens appeared to be in good condition. Fuel swelling is apparent in the slight barrel-like shape of some of the specimens. The cladding over the plenum chambers of all specimens was bright and shiny, while that over the fueled regions had a frosted or etched appearance. This condition appears to be a result of sigma formation in the 306 cladding alloy.

Helium leak testing revealed that two of the specimens had cladding penetrations. The leak in one specimen (836-9) was located in a tack weld at the plenum end of the sample, where a small metal tab had been attached for specimen orientation. However, it appears that this tab

R-48951

SPECIMEN 836-1

R-48942

ORNL-DWG 69-13205

SPECIMEN 336-5

R-48945

SPECIMEN 836-9

R-48936

SPECIMEN 336-2

R-48939

SPECIMEN 554-13

R-48948

SPECIMEN 554-14

R-48933

SPECIMEN 554-12

R-48955

SPECIMEN 554-4

Fig. 5. Posttest Photographs of Fuel Specimens from the ORRF-5 Irradiation Program.

was broken off during disassembly, and thus this particular specimen did not leak during irradiation. The leak in the other specimen (836-1) occurred at the site of a whitish deposit on the outer surface of the sample (see Fig. 6). Gamma spectroscopy subsequently showed this whitish material to contain significant quantities of ^{137}Cs and ^{95}Zr - ^{95}Nb , thus indicating that this probably was at the site of the in-reactor failure.

Fig. 6. Suspected Failure Area on Specimen 836-1. Approximately 2X.

All eight specimens were gamma scanned to determine the axial distribution of fission products within the specimens. We stacked the specimens end to end (separated by 1/4-in.-thick spacers) in a thin-walled aluminum tube and slowly drove the tube past a 0.010- x 1.00-in. collimator slit in the shielding of a NaI scintillation detector. The detector circuitry was adjusted to record only gamma rays with energies above 0.4 MeV. As shown in Fig. 7, scans showed no unusual effects except for strong activity peaks at the specimen plenum chambers. Gamma spectroscopy showed ^{131}I , ^{134}Cs , ^{136}Cs , and ^{137}Cs to be the predominant source of activity in the plenums of these specimens; however, some ^{95}Zr - ^{95}Nb was also detected in plenum chambers of specimens 554-4, 554-13, and 554-14.

The length, inner and outer diameters, and overall volume of the specimens were measured before and after irradiation to determine the dimensional stability of the specimens. Specimen lengths and outer

Fig. 7. Axial Gamma Scans of ORRF-5 Specimens.

diameters were measured with conventional micrometers. The specimen volumes were determined by immersion density techniques. The inner diameters of the specimens were measured with a Compter gage before irradiation; however, a series of go no-go gages were used for the post-irradiation measurements. The overall changes in the specimen dimensions are summarized in Table 2.

The quantity of fission gas released by the specimens during irradiation was determined by puncturing the specimen cladding with a laser and determining the volume and composition of the released gases. The total quantity of fission gas produced during irradiation was then calculated from the analytically determined specimen burnup values, and the fraction of fission gas released was determined from the ratio of these two values. The gas release data are summarized in Table 3. Since specimens 836-1 and 836-9 contained leaks, we could not determine fraction of gas release in these specimens. In addition, the gas samples from specimen 554-12 contained extremely large quantities of air. This indicates that the gas sampling system leaked during the penetrating operation. Consequently, the low indicated gas release fraction for specimen 554-12 is not thought to be realistic.

The end caps of specimens 336-2, 336-5, 554-4, 554-14, and 836-9 were removed to permit inspection of the plenum chambers. As shown in Fig. 8, the plenums of specimens 336-2 and 336-5 were discolored by a relatively light soot-like deposit. However, massive deposits of bright, metallic-appearing crystals were found in the chambers of specimens 554-4, 554-14, and 836-9. These crystals appeared to be somewhat preferentially deposited upon the outer surface of the inner tube diametrically in line with the specimen vent holes.

A transverse cross section of each fuel specimen was prepared for metallographic examination. These sections were examined microscopically for evidence of microstructural changes and/or mechanical damage. Composite photomicrographs of the complete cross section of each specimen are shown in Figs. 9 through 11. The microstructure from the inner cladding to the outer is shown at high magnification in Figs. 12 through 14. Indications of core-cladding separation, fuel redistribution, mass transfer between cermet and cladding, and metallic deposits are evident

Table 2. Summary of Dimensional Changes in the ORRF-5 Irradiation Test Specimens

Specimen	Total Void Volume (%)	End Plenum Volume (%)	Specimen Burnup (fissions/cm ³)	Average Specimen Temperature (°C)	Maximum OD Increase (%)	Maximum ID Decrease (%)	Length Increase (%)	Fuel-Core Volume Increase ^a (%)
			$\times 10^{20}$					
336-2	31	29	1.23	1578	3.6	4.5	0.45	8.95
336-5	30	28	1.23	1505	1.3	2.5	0.44	3.76
554-4	16	7	1.28	1561	2.7	2.3	0.39	6.33
554-12	9	0	1.39	1585	6.7	4.7	0.78	17.12
554-13	10	0	1.28	1549	4.8	2.7	0.51	11.44
554-14	19	9	1.27	1524	1.7	1.6	0.45	4.23
836-1	16	0	1.35	1591	5.0	6.2	0.67	12.21
836-9	21	4	1.32	1534	3.2	2.1	0.30	≈ 8.0

^aCalculated value assuming that the increase in total specimen volume is a result of swelling in the fuel core only (i.e., no change in the specimen plenum volumes).

Table 3. Summary of Fission Gas Release in the ORRF-5
Irradiation Test Specimens

Specimen	Average Specimen Temperature (°C)	Total Void Volume (%)	Voids in Cermet Core (cv) (%)	Fuel-Core Volume Increase (%)	Fission Gas Release (%)
336-2	1578	31	2	8.95	53
336-5	1505	30	2	3.76	66
554-4	1561	16	9	6.33	13
554-12	1585	9	9	17.12	1(?)
554-13	1549	10	10	11.44	38
554-14	1524	19	10	4.23	28
836-1	1591	16	16	12.21	a
836-9	1534	21	17	≈ 8.0	a

^aSpecimen leaked.

SPECIMEN 336-2

SPECIMEN 336-5

R-50033

SPECIMEN 554-4

SPECIMEN 554-14

SPECIMEN 836-9

Fig. 8. Plenum Chambers of ORRF-5 Fuel Pins. 4.5X. Reduced 34%.

Fig. 9. Transverse Cross Section of Fuel Pins. 10X. (a) Specimen 336-2: 1.23×10^{20} fissions/cm³ at 1578°C; 2% core and 29% plenum void volume. (b) Specimen 336-5: 1.28×10^{20} fissions/cm³ at 1505°C; 2% core and 28% plenum void volume. Reduced 18%.

Fig. 10. Transverse Cross Sections of ORRF-5 Fuel Pins. Approximately 6.5X. (a) Specimen 554-4: 1.28×10^{20} fissions/cm³ at 1561°C; 9% core and 7% plenum void volume. (b) Specimen 554-12: 1.39×10^{20} fissions/cm³ at 1585°C; 9% core and 0% plenum void volume. (c) Specimen 554-13: 1.28×10^{20} fissions/cm³ at 1549°C; 10% core and 0% plenum void volume. (d) Specimen 554-14: 1.27×10^{20} fissions/cm³ at 1524°C; 10% core and 9% plenum void volume. Reduced 11%.

Fig. 11. Transverse Cross Sections of ORRF-5 Fuel Pins. 10X. (a) Specimen 836-1: 1.35×10^{20} fission/cm³ at 1591°C; 16% core and 0% plenum void volume. (b) Specimen 836-9: 1.32×10^{20} fissions/cm³ at 1534°C; 17% core and 4% plenum void volume. Reduced 17%.

R-50045

Fig. 12. Transverse Cross Sections of ORRF-5 Fuel Pins. (a) Specimen 336-2: 1.23×10^{20} fissions/cm³ at 1578°C; 2% core and 29% plenum void volume. (b) Specimen 336-5: 1.28×10^{20} fissions/cm³ at 1505°C; 2% core and 28% plenum void volume.

R-50049

Fig. 13. Transverse Cross Sections of ORRF-5 Fuel Pins. (a) Specimen 554-14: 1.27×10^{20} fissions/cm³ at 1524°C; 10% core and 9% plenum void volume. (b) Specimen 554-4: 1.28×10^{20} fissions/cm³ at 1561°C; 9% core and 7% plenum void volume. (c) Specimen 554-13: 1.28×10^{20} fissions/cm³ at 1549°C; 10% core and 0% plenum void volume. (d) Specimen 554-12: 1.39×10^{20} fissions/cm³ at 1585°C; 9% core and 0% plenum void volume. Reduced 26%.

Fig. 14. Transverse Cross Sections of ORRF-5 Fuel Pins. (a) Specimen 836-1: 1.35×10^{20} fissions/cm³ at 1591°C; 16% core and 0% plenum void volume. (b) Specimen 836-9: 1.32×10^{20} fissions/cm³ at 1534°C; 17% core and 4% plenum void volume.

to various degrees in the individual specimens. This is briefly summarized in Table 4. Implications of these various effects are analyzed in detail in the following section.

Table 4. Summary of Microstructural Changes in
ORRF-5 Irradiation Test Specimens

Specimen	Irradiation Temperature (°C)	Core-Cladding Separation, %		Cladding- Cermet Mass Transfer	Fuel Reloca- tion	Metallic Deposits
		Outer	Inner			
836-1	1591	100	≈ 70	Yes	Yes	Yes (gap)
554-12	1585	100	≈ 70	Yes	Yes	Yes (gap)
336-2	1578	≈ 90	≈ 70	Yes	Yes	Yes (gap)
554-4	1561	0	0	No	No	Yes (plenum)
554-13	1549	≈ 80	≈ 70	Some	No	No
836-9	1534	0	≈ 70	No	No	Yes (plenum)
554-14	1524	0	0	No	No	Yes (plenum pinholes)
336-5	1505	0	75	No	No	No

EVALUATION OF POSTIRRADIATION EXAMINATION

Relocation of Fuel

Fuel relocation was observed for those capsules that operated at mean temperatures above 1578°C. No fuel relocation was noted in fuel pins with mean operating temperatures of 1561°C and below, although most of these pins were heated to temperatures above 1600°C during some period of the irradiation. Fuel relocation such as observed in Fig. 15 is typical of that observed by Hilbert *et al.*¹⁰ for UO₂-W cermets at

¹⁰R. F. Hilbert *et al.*, Progress on High-Temperature Fuels Technology During August Through October 1969, BMI-1875, pp. 18-22.

Fig. 15. Cladding-Cermet Interface at the Outer Cladding of Specimen 336-2. 200X. (a) As-polished specimen showing some fuel redistribution, vaporization of cladding, and core-cladding separation. (b) Similar location in the etched condition.

1640°C. The amount of fuel relocation observed in GE-NSP capsules appears to be less than observed in BMI's pins operated at higher temperatures. Relocation of fuel to the extent observed in ORRF-5 did not appear to significantly influence the irradiation behavior.

However, an excessive concentration of ceramic at the core-cladding interface could destroy the bond between cladding and cermet. The advantages of fully bonded cermet fuel systems would therefore be negated.

Metallic Deposits

Deposits in the plenum chamber were found in specimen 554-4, as shown in Fig. 16. These deposits appear to be crystalline, and microprobe analysis indicated the composition to be primarily tungsten with perhaps a trace of molybdenum. No indication of uranium or rhenium was found.

Similar appearing deposits were found in the vent holes of specimen 554-14 and around the outer edge of the core in specimen 836-1, as shown in Fig. 17.

Microstructural Changes in the Cladding

As expected, significant quantities of sigma phase were formed in the cladding. Typical examples can be found in Figs. 18 and 19.

Small grain boundary cavities are shown in Fig. 20 and are typical of those found during high-temperature and low-stress creep testing. The cracks were observed in the cladding of specimen 554-12. Although the volume swelling on this specimen was the greatest of all listed, it did not fail, as determined by release of fission products to the coolant.

Gross microstructural changes were observed on the inner surface of the outer cladding of specimens 836-1, 554-13, and 336-2. The cladding had separated from the cermet in each of these pins. Photomicrographs of an unaffected interface are given in Fig. 21, partial reaction as seen in capsule 554-13 is shown in Fig. 22, and extensive reaction as observed in capsule 836-1 is shown in Fig. 23. The microstructural changes appear to be a result of leaching of one or more elements from the W-Re-Mo cladding. The exact species and the conditions leading to

INNER
SURFACE

DEPOSIT

R-49627

OUTER
SURFACE

DEPOSIT

(a)

INNER CLADDING

R-49165

(b)

PARTITION WALL

Fig. 16. Longitudinal Sections Through Specimen 554-4 Showing Buildup of Solid Material in the End Plenum. (a) 20X. (b) Deposit shown near inner surface in (a) at 200X.

Fig. 17. Metallic Deposits Formed in Fuel Specimens. 200x.
(a) Pinholes of specimen 554-14. (b) Outer edge of the core of specimen 836-1.

Fig. 18. Typical Microstructures of Cladding-Cermet Interface on the Inner Cladding of Specimen 336-2. 200x. (a) As polished. (b) Etched.

Fig. 19. Microstructure of Cladding on Specimen 336-2 at a Location Slightly away from the Failure Point. The formation of sigma phase and limited vaporization on the cermet-cladding interface are seen. 200x. Etched.

Fig. 20. Cladding Cracking in Specimen 554-12. 100x. As polished.

(a)

(b)

Fig. 21. Unaffected Cermet-Cladding Interface on the Inner Surface of the Outer Cladding. 200X. (a) As polished. (b) Etched.

Fig. 22. Start of Cladding-Cermet Reaction on the Outer Cladding of Specimen 554-13. As polished. (a) 200x. (b) 500x.

Fig. 23. Extensive Microstructural Changes in Outer Cladding at Cermet-Cladding Interface (Specimen 836-1). (a) 200x; etched. (b) 500x; as polished.

this phenomenon have not been identified. However, the fact that crystals of metallic tungsten were found in other regions of these capsules suggests that tungsten is being preferentially lost from the cladding and deposited elsewhere. It is perhaps even more important to note that the material is being transported up a temperature gradient, from the cladding to the fuel core, the inner surface of the pinhole vents, and the plenum chamber. This suggests that tungsten is being transported as a chemical compound that vaporizes at some lower temperature and decomposes at a higher temperature. Such an effect could be due to fission-product iodine. During irradiation, iodine could diffuse to the cladding surfaces and form tungsten iodide. The tungsten iodide would vaporize and diffuse back to the higher temperature regions, where it would decompose and form metallic tungsten crystals and free iodine. This iodine would be available to repeat this transport cycle. Such reactions could prove to be a serious problem in all high-temperature fuel clad with tungsten. Additional work will be necessary to confirm or refute this explanation.

Cermet Core-Cladding Separations

Some indication of core-cladding separation was observed on either the outer or inner cladding for all pins except 554-4 and 554-14. Cladding vaporization was observed in all pins showing core-cladding separation except pins 836-9 and 336-5. The latter pin had the highest fission gas release and hence may have been under the highest internal pressure, but it was also the coldest capsule. Also, the separation at the inner cladding could have occurred before irradiation.

Fission Gas Retention

Only those fission products that could be evacuated from the core at room temperature were measured and analyzed. Hence, those products that were present in the tungsten matrix, UO_2 and $\text{UO}_2\text{-ThO}_2$ fuel agglomerates, and in the W-Re-Mo alloy cladding were not directly determined.

The fission gas release as a function of mean irradiation temperature is plotted in Fig. 24(a). Since fission gas data are not available from any of the 836 type capsules, one can only compare the data from the 336 series to the three capsules in the 554 series. Regardless of irradiation temperature, the 554 capsules containing 9.2 wt % ThO₂ in the ThO₂-UO₂ agglomerates showed less gas release than the 336 specimens, which do not contain thorium. This relationship is shown in Fig. 24(c), which suggests that gas release from the 836 series would have been the lowest of all capsules.

Newsom and Collins¹¹ reported earlier data for W-UO₂ cermet. At a burnup of 6.7×10^{19} fissions/cm³ the gas release was 24% of the total generated. However, at higher burnup, mobile fission gases increased to 50%. Thus, the values for capsules 336-5 and 336-2 of 66 and 53%, respectively, appear to be reasonable.

Further interpretation of these data cannot be made without total fission product inventory, which was beyond the budget for the program.

Effectiveness of Void Deployment

Comparison of the volume swelling data of all the capsules in the 554 series indicates that both irradiation temperature and total void content are very important but that cermet cores with 10% porosity do not need to have vents to make the end plenums effective. Capsule 554-14 with vents swelled less than 554-4 without vents; however, the latter pin operated at a higher temperature. Using Fig. 25(a) to estimate the influence of temperature, the importance of vents seems small.

The swelling behavior of the 336 series indicates a similar dependency on irradiation temperature and total void content. For example, as shown in Fig. 25(b), the 30% void volume available in 336-2 appears to effectively reduce swelling when compared to 836-1 and 554-12, specimens which contained only 16 and 9% porosity, respectively. These pins operated at nearly the same temperature but contained differing amounts

¹¹D. L. Newsom and J. F. Collins, Irradiation Testing and Evaluation of Refractory Metal Fuel Elements of 710 Fast Reactor Program, GEMP-710 (October 1969).

Fig. 24. Fission Gas Release as a Function of (a) Mean Irradiation Temperature, (b) Cermet Void Volume, and (c) Thoria Content.

operated at very nearly the same mean temperature. Their swelling data are plotted in Fig. 26. Capsules 554-13, 554-4, and 554-14, which had equal cermet porosity, are plotted in Fig. 27. While we appreciate the uncertainties associated with curves drawn with so few data points, the curves seem to make sense. From the two curves, one gathers that voids within the core or in the end plenum are effective to nearly the same extent of 70 to 80%. Having some confidence in the data, one could suggest that 24 to 33% void volume is needed to reduce the swelling to zero for this high-temperature range. Recently, Hilbert et al.¹³ at BMI reported negative swelling for a $\frac{1}{2}$ - UO_2 cermet irradiated at 1640°C to

¹³R. F. Hilbert et al., Progress on High-Temperature Fuels Technology During August Through October 1969, BMI-1875, pp. 18-22.

Fig. 26. Effect of Core Voids on Composite Swelling.

Fig. 27. Effect of End-Plenum Voids on Composite Swelling.

1.7×10^{20} fissions/cm³. That cermet contained 28 vol % porosity. The data in Fig. 27 would suggest that void volume in the end plenum is only effective up to 14%. One can imagine several phenomenon that would support an upper limit on the effective end plenum void. Considering this possibility, Fig. 25(b) can be redrawn to show swelling versus effective total void volume. These values are shown in Fig. 28. If the deduction is correct, then total void volume of 24 to 33%, either all within the core or 14% void volume in the end plenum with the remainder in the core, will reduce the swelling to a minimum value.

Evaluation of the data from all these capsules was difficult because of the many variables, particularly if one believes that irradiation temperature over the range of 1500 to 1600°C is an important factor. Even so, these data certainly support those who believe that deployment of voids in cermets containing large volume fractions of ceramic fuel will accommodate the fission products at irradiation exposures up to 2×10^{21} neutrons/cm³.

Fig. 28. Composite Swelling vs End-Plenum Voids.

Cause of Fuel Capsule Cladding Failure

Two capsules leaked during our hot-cell examination. As explained earlier, the failure in 836-9 was attributed to removal of a small metal tab during disassembly. But the other capsule, 836-1, represents the pin that failed in-reactor. Noting that this pin failed at a diametral swelling lower than that sustained by capsule 554-12, we believe this failure to have been premature. The early failure is attributed to the extensive mass transfer observed on the inner surface of the outer cladding that we discussed earlier. If this reaction seriously reduces the load-carrying ability of the cladding, the unaffected portion of the cladding will fail in a ductile manner. The cladding failure in capsule 836-1 is shown in Fig. 29. The failure appears to be both inter- and intragranular. The reaction that occurred in the cladding was probably the primary cause of this cladding failure. Without more knowledge than is now available concerning this reaction, it may be difficult to control in the future.

CONCLUSIONS

From the postirradiation examination and evaluation, we conclude the following:

1. The irradiation performance of the refractory metal cermets depended on their mean operating temperature in-reactor and thei

Fig. 29. Cladding Microstructure of Specimen 836-1 Near Failure Point. Note unusual microstructure on the outer surface of the cladding in addition to the extensive reaction at the cermet-cladding interface. 200x. Etched.

total as-fabricated void space. The most dimensionally stable specimens operated at the lowest temperatures and contained the largest void volume. Greatest swelling and evidence of actual and incipient failures occurred in specimens subjected to the highest temperature and containing the least void volume.

2. The total void space of the refractory metal cermets could have been located in the fuel dispersion, up to 14% in the end plenum chambers, or distributed between these two regions with equivalent effectiveness. Aside from a means of increasing total void volume, drilling small holes into the fueled region of the cermet (to shorten the diffusion path of the permeable fission gases) was not necessary for these cermets having at least 2% porosity within the core.

3. The maximum total void volume that appears to effectively accommodate fission products by reducing swelling is 24 to 33 vol %, but the maximum effective void volume in the end plenum is 14%.

4. Reactions at the cladding-cermet interface reduce the mechanical integrity of the cladding and can cause premature failure. The

cause is not known and needs to be investigated. This serious effect may be due to the formation of tungsten iodide from fission products and needs to be investigated further.

ACKNOWLEDGMENT

The authors wish to acknowledge the assistance of J. A. McGurty, J. F. Collins, and D. L. Newsom from GE-NSP and H. Reesor, D. Gates, L. L. Allen, and E. D. Sims from the Metals and Ceramics Division of ORNL for their assistance during this examination.