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FUELS COMMITTEE MEETING - MAY 19-21, 1964.

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FUELS COMMITTEE MEETING - May 19, 20, 21, 1964

G. A. Last

SUMMARY

Metallic thorium uranium fuel elements continue to show excellent irradiation performance in high temperature pressurized water coolant. Volume expansion measurements made after 2.1×10^{20} fissions/cm³ (6200 MWD/T) at fuel temperatures above 500 C shows no indication of fission gas-induced swelling.

Analysis of fuel swelling data from tubular elements shows an apparent effect of geometry (restraint) on both low temperature volume expansion and the temperature at which accelerated volume expansion initiates. Evidence of grain boundary tearing has been observed in tubular metallic uranium fuels irradiated in 1600 psi water coolant.

Volume expansion due to two types of structural damage have been observed in uranium - 2 w/o zirconium alloy irradiated to 0.25 a/o burnup at temperatures up to 550 C.

Irradiation Testing of Thorium-Uranium Fuel Elements - R. K. Marshall,
J. W. Goffard

Irradiation testing of three tubular thorium-uranium fuel elements in the high-temperature, high-pressure, water-cooled ETR P-7 facility continued. The test elements are Zircaloy-2 clad Th - 2.35 w/o U-235 - 1 w/o Zr alloy fuel 4.45 cm (1.75 inch) O.D. x 2.67 cm (1.05 inch) I.D.

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The test facility has operated at 204 C (400 F) coolant inlet at a pressure of 110 atmospheres (1600 psi). The central element, operating at the highest power, has accumulated a maximum exposure of 2.1×10^{20} fissions/cm³ (6200 MWD/T). Maximum operating conditions and measured volume changes for the irradiation period are given in Table I.

As seen in Table I, the measured fuel swelling is low, the 0.8 percent volume increase observed after cycle 62 irradiation is approximately equal to the theoretical minimum volume expansion due to fission product production. The measured volume expansion for the two lower power test elements at the end of cycle 62 was 0.8 and 0.6 percent.

Visual examination of the elements has indicated excellent fuel performance throughout the irradiation. The fuel elements have remained free of films and deposits with no evidence of accelerated corrosion on the cladding surfaces or in the braze closure areas.

Submicron UC Dispersion in Metallic Uranium - R. K. Marshall, J. W. Weber

Metallic uranium fuel rods containing a submicron dispersion of uranium carbide are being irradiated to establish the effect of carbide size on irradiation stability of metallic uranium.

A NaK capsule containing two uranium rods one fine and one with coarse uranium carbide dispersions was charged into the ETR in December, 1963. These fuel rods have accumulated an exposure of 0.20 atom percent burnup; goal exposure for this capsule is 0.3 atom percent burnup. The maximum fuel temperature is 550 C. Following capsule discharge in July these rods will be subjected to metallographic evaluation and post-irradiation annealing studies.

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Table I - IRRADIATION HISTORY OF TUBULAR THORIUM -
2.35 w/o U-235 - 1.0 w/o ZIRCONIUM FUEL ELEMENTS

ETR Cycle	BU Fissions/cm ³ (MWD/T)	Temp °C	Maximum Operating Conditions		
			Surface Heat Flux cal/sec-cm ² (BTU/hr-ft ²)	Specific Power watts/gm (kw/ft)	Fuel Volume Increase, Percent
54	2.8x10 ¹⁹ (800)	585	75 (1.0x10 ⁶)	69 (210)	0.15
55	5.4x10 ¹⁹ (1550)	538	68 (9.0x10 ⁵)	62	0.15
56	8.6x10 ¹⁹ (2475)	523	65 (8.6x10 ⁵)	59 (180)	Not Weighed
57	1.2x10 ²⁰ (3400)	585	75 (1.0x10 ⁶)	69 (210)	0.5
58	Not irradiated	-	-	-	-
59	1.2x10 ²⁰ (3500)	460	52 (6.9x10 ⁵)	46 (141)	0.5
60	1.6x10 ²⁰ (4500)	540	65 (8.7x10 ⁵)	60 (183)	0.5
61	1.8x10 ²⁰ (5200)	540	65 (8.7x10 ⁵)	60 (183)	0.8
62	2.1x10 ²⁰ (6200)	520	62 (8.3x10 ⁵)	57 (174)	0.8

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Irradiation Performance of Tubular Fuel Elements - J. W. Goffard

Additional swelling and operational data have been obtained from two KER charges of N-Reactor fuel elements. The N-Reactor fuel element (NAE) consists of an inner (NIE) and an outer (NOE) tubular component. The charges were irradiated to tube average exposures of approximately 1600 and 2000 MWD/T at prototypic temperatures and 1600 psi coolant pressure. Maximum fuel swelling observed in the 1600 exposure test was 0.85 v/o in an NIE (@ 2000 MWD/T) and 2.50 v/o in the corresponding NOE (@ 2500 MWD/T). Maximum fuel swelling observed in the 2000 exposure test was 1.33 v/o in an NIE (@ 2200 MWD/T) and 3.64 v/o in an adjacent NOE (@ 3100 MWD/T). This new data along with previously obtained N fuel component data is portrayed graphically in Figure 1.

Analysis of the N-Reactor fuel data (110 points) and special single tube element data (KSE-3 and KSE-5, 20 points) that operated to maximum exposures of 3600 MWD/T and as high as 535 C vol. mean fuel temperature, has resulted in three similar expressions for the fuel swelling - one for each fuel component geometry. The expressions are -

for NOE,

$$\% \Delta V \text{ in fuel} = B \left[4.15 + 550 \left(\frac{T_r}{P_r} \right) \left(\frac{1}{1+e^{-\frac{T+100B-420}{20}}} \right) \right]$$

for NIE,

$$\% \Delta V \text{ in fuel} = B \left[3.05 + 550 \left(\frac{T_r}{P_r} \right) \left(\frac{1}{1+e^{-\frac{T+100B-480}{20}}} \right) \right]$$

for KSE 3&5,

$$\% \Delta V \text{ in fuel} = B \left[3.95 + 550 \left(\frac{T_r}{P_r} \right) \left(\frac{1}{1+e^{-\frac{T+100B-460}{20}}} \right) \right]$$

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where,

$B = \text{a/o burnup, (1 a/o } \approx 8650 \text{ MWD/T)}$

$T_r = \frac{\text{vol. mean fuel temp., } ^\circ\text{K}}{273}$

$P_r = \text{restraining pressures from outer clad and coolant}$
(on basis of 20,000 psi Zr-2 strength)

$T = \text{vol. mean fuel temp. } ^\circ\text{C}$

Both the minimum swelling rate (first term inside of the brackets) and the initiation of accelerated volume expansion (exponential term) vary with the three tubular geometries studies.

In Figure 2, the experimental or measured fuel volume changes are compared with the volume changes obtained with the above expressions.

Radiometallurgical examination is being conducted on an NOE which was irradiated to 3100 MWD/T and which experienced 3.64 percent volume expansion. Superficial examination has revealed no deficiencies in the performance of the NOE; i.e., no obvious warp, no bumping or rippling of the surfaces, no crud deposits or corrosion effects, and no inner bore instability. Metallographic examination of the uranium fuel with the light microscope to 1000X reveals no spherical porosity such as might be generated by gas agglomeration. However, the examination does show the presence of small voids in the hashed or swirled microstructures. Except for their small size, these voids are similar to the grain boundary tearing voids which are observed in uranium irradiated at approximately 400 C. Electron microscopy will be used to further study the structure and to determine the source of the fuel swelling.

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Swelling Characteristics of Uranium and Uranium Alloys - J. W. Weber

Irradiation studies are underway on a series of uranium alloys with various combinations of additions of Fe, Si, Al, Zr, Nb, Mo, and carbon to determine the influence of the alloy additions on the fuel swelling. Four primary causes of volume increase have been observed metallographically.

- 1) Fission gas porosity.
- 2) Cracks both inter- and intra-granular.
- 3) Tearing which results in irregularly shaped holes at grain boundaries and other boundaries where there have been large plastic deformations.
- 4) Pores, which may be round or irregularly shaped, that are aligned in parallel rows, the rows being perpendicular to the long axis of twins in the surrounding uranium.

The fourth phenomenon was first observed and reported by Leggett and Bierlein.⁽¹⁾

As part of the alloy investigation program, a rod of U+2 w/o Zr coextrusion clad with Zircaloy-2 was irradiated to approximately 0.25 a/o (1.2×10^{20} f/cm³) burnup at a calculated maximum temperature of 550 C. Preirradiation heat treatment consisted of holding for ten minutes in the $\beta + \gamma_1$ region at 730 C, quenching into salt at 300 C, holding for five minutes then quenching in water.

Figure 3 shows a typical irradiated microstructure in the center of the rod (550 C) which is different than any previously seen in irradiated

(1) HW-77954, Quarterly Progress Report, Metallurgy Research Operation, April, May, June, 1963.

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U-2 w/o Zr alloy. The appearance of the many large irregularly shaped pores aligned perpendicular to the major axis of the twins is very similar to that observed in unrestrained thin cylinders of uranium.⁽¹⁾ This is the fourth type of structural change mentioned above. Moving radially from the center toward lower temperatures, the number and size of these pores diminishes. At approximately 500 C, all evidence of the aligned pores has disappeared. Between 500 C and 475 C there is a zone almost devoid of pores. Below 475 C the uranium alloy exhibits tearing at grain boundaries and deformation boundaries. This is the third type structural change mentioned before. These tears, however, do not have the same aligned relationship as is seen in the center of the rod. The uranium in this colder region takes on a very distorted appearance and very few clear microstructural features can be seen. Figure 4 shows a typical structure of the fuel at a temperature of approximately 450 C.

At the Sixteenth High Temperature Fuels Committee Meeting it was reported⁽²⁾ that swelling of U-2 w/o Zr rods coextrusion clad with Zircaloy-2 was several times that of unalloyed uranium for comparable temperatures and exposures. Since then, additional experimental data has necessitated a recalculation of the temperature distribution in the rods. The results show a maximum irradiation temperature approximately 200 C higher than was previously calculated and the data now shows that the swelling of the alloyed fuel to be approximately 30 percent higher than unalloyed uranium at equivalent temperatures and burnups rather than the 200 to 300 percent previously reported.

(2) HW-77064, Hanford Contribution for the Sixteenth High Temperature Fuels Committee Meeting, May 1963, G. A. Last, (Confidential.)

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These higher temperatures have been substantiated metallographically. Work by Lagerberg⁽³⁾ on U-2 w/o Zr and by Zegler⁽⁴⁾ on U-4 w/o Zr show a microstructure obtained by annealing - 1 day and 1 week respectively - at 690 C, which is identical to that observed near the center of the 0.25 a/o burnup rods. From an interpretation of microstructures, the center of the rod is now believed to have been in the $\beta + \gamma_2$ temperature range during irradiation.

Irradiation Performance of Fluted Tubular Fuel Element - D. P. O'Keefe

One design concept which is being investigated as a means of extending the total exposure capability of metallic uranium fuel is a fluted outer surface for tubular elements. The advantage sought in this design is a surface which can accommodate volume increases of the core without the generation of tensile strain in the cladding.

Two N-Reactor size single tube elements embodying this design are being irradiated in the M-3 high pressure water loop in the ETR. The outer surface of these elements has eight flutes 0.1 inch high which are generated by circular arcs. This shape will permit, geometrically, a volume expansion of 11 percent without appreciable tensile elongation of the cladding.

-
- (3) Lagerberg, G., Phase Transformations in a Uranium Zirconium Alloy Containing 2 w/o Zirconium. Jour. of Nuc. Mat. 9 No. 3, 1963.
- (4) Zegler, S. T., The Uranium Rich End of the Uranium-Zirconium System, ANL-6055, 1962.

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The elements are now undergoing their eleventh cycle of reactor operation in the EFR. They have been examined after each cycle, and after ten cycles appeared to be in excellent condition. After the tenth cycle a slight flattening was visible in the bottom of the flutes. This shape change, caused by expansion of the fuel, was uniform for all of the flutes and is the expected change as swelling progresses. Table II shows the irradiation history and volume expansion for the elements of this test.

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Table II - IRRADIATION HISTORY OF N-REACTOR SIZE FLUTED FUEL
ELEMENT

ETR Cycle	BU Fissions/cm ³ (MWD/T)	Temp °C	Outer Surface	Specific	Fuel Volume	
			Heat Flux cal/sec-cm ² BTU/hr-ft ²	Power watts/gm (kw/ft)	Increase Percent Element 10-67	Element 10-68
52	1.1x10 ¹⁹ (200)	520	38 (505,000)	35 (130)	0	0
53	2.3x10 ¹⁹ (400)	"	"	"	0	0
54	3.5x10 ¹⁹	"	"	"	.2	.3
55	4.5x10 ¹⁹	"	"	"	.2	.4
56	5.7x10 ¹⁹ (1000)	"	"	"	.2	.5
57	6.8x10 ¹⁹ (1200)	"	"	"	.4	.7
58	7.9x10 ¹⁹ (1400)	450	29 (388,000)	27 (100)	.7	.9
59	9.0x10 ¹⁹ (1600)	450	28 (388,000)	"	.7	1.1
60	1.0x10 ²⁰	450	"	"	.7	1.1
61	Not irradiated		-	-	-	-
62	1.13x10 ²⁰ (2000)	550	41 (544,000)	38 (140)	1.3	1.9

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FIGURE 1

Fuel Swelling in Four Changes of N-Reactor Fuel Elements

FIGURE 2

Comparison of Measured Fuel ΔV with Calculated ΔV Using
Appropriate Expression for each Geometry Type

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1800X

3000X

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FIGURE 3

Uranium - 2 w/o Zirconium Irradiated to 0.25 w/o Burn-up at 550 C
All Pictures are near the Center of the Rod

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As Polished

250X

Cathodic Etch

250X

1800X

3000X

Electron Photomicrographs

FIGURE 4

Uranium - 2 w/o Zirconium Irradiated to 0.25 w/o Burn-up at 450 C
All Pictures are near the Edge of the Rod

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