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RESEARCH AND DEVELOPMENT REPORT

HWA-74941

FINAL REPORT

IRRADIATION PERFORMANCE

ENRICHED SEVEN-ROD GUSTAFSON ELEMENT

WITH 20- AND 30-MINUTE COOLING

SUPPLEMENT B TO PRODUCTION OF THE ELEMENT

STANFORD LABORATORIES

STANFORD ATOMIC ENERGY LABORATORY
RICHLAND, WASHINGTON

GENERAL ATOMIC CORPORATION

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Materials
(M-3679, 28th Ed.)

FINAL REPORT:
IRRADIATION PERFORMANCE
OF ENRICHED SEVEN-ROD CLUSTER FUEL ELEMENTS
WITH 20- AND 30-MIL-THICK CLADDING,
SUPPLEMENT B TO PRODUCTION TEST IP-226-A (RM-562)

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By

R. L. Call

~~By Authority of~~

Metallurgy Development
Reactor and Fuels Laboratory
Hanford Laboratories

July 1962

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By Authority of CG-PR-2
RM 562, 3-10-94
Chris Males, 3-31-94
Verified by DK Hanson, 4-4-94

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FINAL REPORT:
IRRADIATION PERFORMANCE
OF ENRICHED SEVEN-ROD CLUSTER FUEL ELEMENTS
WITH 20- AND 30-MIL-THICK CLADDING,
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INTRODUCTION

The proposed operating conditions for Hanford's N-Reactor fuel elements require the fuel to be an extended surface geometry and to maintain adequate mechanical strength and corrosion resistance in high-temperature water. The fuel element geometry which has been selected for N-Reactor use is a Zircaloy-2-clad, coextruded, tube-in-tube fuel element. A seven-rod cluster fuel element had been considered early in the N-Reactor design program. This report describes the testing of seven-rod cluster fuel elements fabricated from coextruded Zircaloy-2-clad 1.6% enriched uranium rods and compares the effects of two cladding thicknesses, 20 mils and 30 mils, on the behavior of the uranium. The results obtained from this test, although not directly applicable to N-Reactor fuels, show the irradiation performance of Zircaloy-2-clad uranium in high temperature water.

SUMMARY AND CONCLUSIONS

Six seven-rod cluster fuel elements were irradiated to a total exposure of 2520 Mwd/ton in high-temperature water. The fuel elements were fabricated from coextruded, 0.592-inch diameter, Zircaloy-2-clad rod stock. The rods of four of the clusters had 20-mil-thick cladding; the others had 30-mil-thick cladding. The test was terminated before goal exposure was reached when rupture signals were received by rupture monitors on the irradiation test facility. A peripheral rod of one of the clusters having 20-mil-thick cladding was found to have a split in the cladding. The failure is attributed to localized plastic straining and eventual failure of the cladding as the uranium swelled during irradiation.

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The rods of the cluster with the failure and the rods of one of the clusters with 30-mil-thick cladding were measured following irradiation. The average diameter increase of the rods with 20-mil-thick cladding was 0.0005 inch and of the rods with 30-mil-thick cladding, 0.0029-inch.

A previous test of seven-rod clusters⁽¹⁾ failed at about the same exposure; here also the failure was a split in the 20-mil-thick cladding of a peripheral rod. In this earlier test, the rods with 20-mil-thick cladding increased 0.005 inch in diameter while the rods with 30-mil-thick cladding increased 0.0020 inch in diameter. Thus, fuel elements with 20-mil-thick Zircaloy-2 cladding can fail by localized plastic straining of the cladding even though the overall strain is very low. No evidence of failure was observed in the fuel rods with 0.030-inch cladding.

DISCUSSION

A. Fabrication of Coextruded Rod Stock

The coextruded rod stock from which the fuel elements were made was obtained from Nuclear Metals, Inc. (NMI) of Cambridge, Massachusetts. The rods with 20-mil-thick cladding were made from NMI extrusions No. 18327, 18329, 18330, 18331, and 18332. The rods with 30-mil-thick cladding were made by cutting up 36-inch long integrally end closed coextruded rods. The integrally end closed rods used were No. 5, 6, 17, 20, 30, 33, 34, 35, 36, 37, 38, and 39. The complete details of the coextrusion process are given by Lowenstein.⁽²⁾ A brief description is given below.

1. Machine uranium ingots to size.
 2. Double beta heat treat.
 3. Clean uranium and Zircaloy-2 sleeves and insert uranium into sleeves.
 4. Insert these assemblies into copper cand, evacuate, and seal.
 5. Coat assembly with graphite suspended in alcohol, heat, and extrude.
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- 6. Remove copper.
- 7. Swage to final size of 0.593 inch.

B. Fabrication of Fuel Elements

The process for fabricating the seven-rod cluster fuel elements from the coextruded rod stock is given below:

- 1. Cut rods to length.
- 2. Machine uranium from the ends of the rods to a depth of 0.300 inch.
- 3. Pickle rod ends in hot nitric acid to remove residual uranium from the inside surface of the cladding in the counterbored ends.
- 4. Weld Zircaloy-2 end caps into counterbored ends using the electron beam welding process.
- 5. Beta heat treat rods at 710 C (1310 F) in trichloride salt for 5 minutes and air cool.
- 6. Machine slots into end caps to receive the end supports.
- 7. Mechanically straighten warped rods.
- 8. Pickle rods in a HNO_3 -HF solution and autoclave for 72 hours in 400 C (752 F), 1500 psi steam.
- 9. Test integrity of cladding-core bond layer by ultrasonic equipment.
- 10. Measure rod diameters.
- 11. Weld rods to the end supports using the inert gas arc welding process.

The fuel elements irradiated in this test are shown in Figure 1.

C. Irradiation History

The six fuel elements shown in Figure 1 were charged into a high temperature, high pressure, recirculating water facility in a Hanford reactor. They were irradiated under the provisions of Supplement B to

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FIGURE 1

Seven-Rod Cluster Fuel Elements Prior to Irradiation
~0.3X

059605-2
REF ID: A60110

Production Test IP-226-A.⁽³⁾ The loop cooling water was maintained at high temperature and pressure during the test and pH was maintained at 10.0 by flow through a lithium-base ion bed and by LiOH additions. The test was terminated after the fuel elements had received an average total irradiation exposure of 2520 Mwd/ton when rupture signals were received from the test facility.

After the fuel elements were discharged, one of the peripheral rods of a cluster made up of rods having 20-mil-thick cladding was found to be ruptured. The volume-average uranium temperatures of the rods of the cluster with the rupture and of the rods of a cluster with 30-mil-thick cladding are given in Table I.

TABLE I

VOLUME-AVERAGE CORE TEMPERATURES

Ruptured Cluster (20-mil-thick cladding)

Peripheral rods	404 C (759 F)
Center rod	369 C (697 F)

Cluster with 30-mil-thick cladding

Peripheral rods	396 C (745 F)
Center rod	342 C (648 F)

D. Postirradiation Examination

The cluster with the rupture and one of the clusters with 30-mil-thick cladding were examined in a radiometallurgy cell. The rupture was a longitudinal split in the cladding of a peripheral rod of the cluster. The ruptured cluster is shown in Figure 2. Below the lower end of the split in the cladding is a striation. This striation extends downward for approximately 1 inch, is missing for a half inch, and then continues on for approximately 1-1/2 inches. The numerous scratches which can be seen on the surfaces of the rods were made during the postirradiation handling. The white spot on the ruptured rod about one-third the way up from the bottom was made

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FIGURE 2

The Ruptured Cluster "As Received"
in a Radiometallurgy Cell
~0.6X

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when a piece of Tigon tubing came in contact with the thermally hot rod in the radiometallurgy cell. Except for scratches made during handling, the black autoclave film on the rods was intact and there was no crud film visible. Many small bumps in the cladding are visible in Figure 2, however.

Both the ruptured cluster and the cluster with 30-mil-thick cladding were disassembled and individual rod diameter measurements were made. Diameters were measured 1-1/2 and 4 inches from each end and at the center. At each place two measurements were made 90 degrees apart. The average pre- and postirradiation diameters are given in Table II. Also given in Table II are the pre- and postirradiation density measurements and the results of the chemical burnup analysis.

TABLE II
PRE- AND POSTIRRADIATION MEASUREMENTS

	<u>Ruptured Cluster</u>	<u>Cluster with 30-Mil-Thick Cladding, Inch</u>
Mean Preirradiation Diameter (all rods)	0.5910 in.	0.5900
Mean Postirradiation Diameter (all rods)*	0.5915 in.	0.5929
Mean Diameter Increase	0.0005 in.	0.0029
Mean Preirradiation Ellipticity**	0.0001 in.	0.0002
Mean Postirradiation Ellipticity	0.0015 in.	0.0012
Preirradiation Density***	18.89 g/cc	
Postirradiation Density***	18.21 g/cc	
Density Decrease	0.68 g/cc (3.55%)	
Burnup (of the ruptured rod only)	0.344 at. %	

* Does not include diameter measurements at the rupture.

** Ellipticity is defined as the difference between two measurements 90 degrees apart on a rod.

*** Density measurements were made only on the ruptured rod and on the extrusion from which it came.

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Figure 3 is a sketch of the ruptured rod showing where the rod was cut during the metallographic examination. The locations of the various specimens discussed in the report are also shown. Metallography was done on one or both of the cut faces of pieces 1 through 5. Piece 2 was dissolved for the burnup analysis and piece 6 was de-clad and used for the postirradiation density measurement.

FIGURE 3

Cutting Sketch of the Ruptured Rod

The right face of piece 1 was polished and etched by ion bombardment. After the etch, cracks were seen in the cladding near the cladding-to-core bond layer. The significance of these cracks is discussed in the Appendix.

The left face of piece 2 is shown in Figure 4 in the "as polished" condition. The striation in the cladding can be seen at the top, just to the left of center. Figure 5 shows the cladding in the necked region of Figure 4. In the upper photomicrograph, bright field illumination reveals numerous small voids in the cladding. The voids vary in size and in density with the amount of necking that has occurred. Similar voids were observed by Forscher in zirconium tensile specimens.⁽⁴⁾ The voids shown in Figure 5 are believed to be a result of deformation which occurred in the cladding during irradiation.

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Striation
↓

FIGURE 4

Left Face of Piece No. 2
"As Polished"
~2X

In the lower photomicrograph of Figure 5, polarized light revealed the grain structure of the cladding. The grains at the sides are undistorted and equiaxed while those in the center are elongated as a result of the straining which took place. The fact that deformed grains are visible is additional evidence that the deformation took place in-reactor because the beta heat treatment the rods received during the fabrication process produced an equiaxed grain structure throughout the cladding.

The cladding-to-core interface was irregular in all cross-sections examined. This irregularity gave the cladding a nonuniform thickness. Figure 6 is typical of the cladding in regions away from the necked region and shows the variations in thickness.

The left face of piece No. 3 is shown in Figure 7 in the "as polished" condition. The striation in the cladding is visible at the top center of the figure. Two photomicrographs of the necked area of Figure 7 are shown in Figure 8. The necked area is also shown at higher magnification in the two photomicrographs of Figure 9.

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Bright Field

Polarized Light

FIGURE 5

Cladding in the Necked Area of Figure 4
(Chemical Etch) 250X

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FIGURE 6

Cladding in a Region away from the Necked Area
(Chemical Etch) 100X

14376

FIGURE 7

Left Face of Piece No. 3
("As Polished") ~4X

14156

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Bright Field Illumination

Polarized Light

FIGURE 8

Cladding in the Necked Area of Figure 7
(Chemical Etch) 75X

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Bright Field →

← Polarized Light

FIGURE 9

The Necked Area of Figure 7 at High Magnification
(Chemical Etch) 250X

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Voids and deformed grains similar to those visible in Figure 5 can be seen in the cladding in Figure 9. The voids in Figure 9 are smaller and fewer than those in Figure 5, as might be expected when it is noted that the cladding in Figure 9 has been strained less than the cladding in Figure 5. Twinning is also visible in the grains of Figure 9.

A large crack extended radially inward toward the uranium core from the necked area in the cladding in the right face of piece No. 3 (Figure 10). This crack is also visible in the left face of piece No. 4 (Figure 11). When the ruptured rod was first cut and the crack was seen extending radially inward from the split in the cladding (Figure 11) it was thought that the crack in the uranium core had caused the cladding to fail. When no crack was found in the uranium associated with the necked cladding (Figures 4 and 7), it was concluded that some other mechanism was responsible for the cladding failure.

It is believed that when uranium is being irradiated at high temperature, it becomes highly plastic and behaves somewhat as a highly viscous fluid.⁽⁵⁾ Also, during irradiation, the cladding loses its ability to elongate uniformly and becomes plastically unstable.^(6,7) If the cladding thickness is nonuniform, as in Figure 5, the cladding will then elongate and "neck" at a thin spot as the uranium swells. Thus, a small increase in rod diameter can cause a local necking and finally a failure in the rod.

It is believed that the above mechanism caused a previous seven-rod cluster irradiation test to fail,⁽¹⁾ and this same failure mechanism was also responsible for the failure of the inner tube of a tube-in-tube fuel element after irradiation to 3200 Mwd/ton.⁽⁸⁾

Figure 12 is a photomicrograph of the split in the cladding shown in Figure 11. The necking (reduction in thickness of the cladding) which occurred prior to the failure can be seen. A portion of the radial crack running into the core can also be seen. The light-colored material in the crack is uranium oxide.

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FIGURE 10

Right Face of Piece No. 3
("As Polished") ~5X

14136

"As Polished"

~4X

Etched by Ion Bombardment
~3X

FIGURE 11

Left Face of Piece No. 4

14048, 14051
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FIGURE 12

Large-Scale View of the Split in the Cladding Shown in Figure 11
(Etched by Ion Bombardment) 50X

14052

The right face of piece No. 4 was polished but not etched. Several cracks typical of cracks in uranium in the "as polished" condition were found in this face (Figure 13).

The left face of piece No. 5 is shown in Figure 14. This face is a cross-section through the center of the split and shows the extent of the uranium corrosion. Again, as in the previous seven-rod cluster failure,⁽¹⁾ the corrosion progressed mainly along the cladding-to-core bond layer and the depth of penetration into the core was not very great.

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FIGURE 13

Cracks in the Uranium of the Right Face of Piece No. 4
("As Polished") 50X

14465, 14466

FIGURE 14

Left Face of Piece No. 5
("As Polished") ~ 1.8X

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Figure 15 is a photomicrograph of the break in the cladding of the section shown in Figure 14. The necking which occurred prior to failure is evident.

FIGURE 15

14463 Photomicrograph of Split in the Cladding in Figure 14
("As Polished") 50X

Figure 16 shows the left and right limits of the corrosion penetration and separation of the cladding from the core in the left face of piece No. 5 (see Figure 14 for view of entire face). This figure shows that the failure occurred on the uranium side of the cladding-to-core bond layer.

A second rod of the cluster which had the rupture was sectioned for metallography. A cross section of this rod can be seen in Figure 17. Several cracks were observed in the uranium of this cross section and one of them is shown in Figure 18, both "as polished" and "as etched by ion bombardment". The difference in the appearance of the crack, which is caused by the difference in the surface preparation, can be seen. Some additional cracks which were observed in the uranium at the cladding-to-core bond layer are shown in Figure 19.

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FIGURE 16

The Left and Right Limits of the Separation of the Cladding from the Core
in Figure 12 ("As Polished") 100X

FIGURE 17

Section Through a Nonfailed Rod in the
Cluster with the Rupture Rod
(Etched by Ion Bombardment) ~2X

14158, 14159, 14804
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Etched by Ion Bombardment

"As Polished"

FIGURE 18

Two Views of a Crack in the Uranium
of the Cross Section Shown in Figure 17
50X

FIGURE 19

Cracks in Uranium at Bond Layer of Figure 17
("As Polished") 50X

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The cluster made up of rods having 30-mil-thick cladding is shown in Figure 20. The rods of this cluster were clean and free from film. No warping of the rods was seen and the rod surfaces were smooth and free from defects. A peripheral rod of this cluster was sectioned for metallography. Cracks, similar to those discussed above, were seen in the uranium of the sectioned rod.

Replicas were made of several of the cross-sections of the ruptured rod. These replicas were prepared for electron microscopy. The electron micrographs revealed nothing unusual. Typical electron micrographs of the cladding, cladding-to-core bond layer and of the uranium core are shown in Figures 21, 22, and 23, respectively.

ACKNOWLEDGEMENT

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FIGURE 20

Cluster with 30-Mil-Thick Cladding
~0.6X

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392-D, 392-A
SECRET

FIGURE 21
Electron Micrograph of the Cladding
of the Ruptured Rod
5000X

Uranium †

FIGURE 22
Electron Micrograph of the Cladding-
to-Core Bond Layer of the Ruptured Rod
10,000X

Zircaloy-2
↓

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FIGURE 23

Electron Micrograph of the Uranium Near the Center
of the Rupture Rod
5000X

396C

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APPENDIX

As part of the metallographic examination of the ruptured rod, some of the cut faces were etched in vacuum by ion bombardment. When the right face of piece No. 1 (see Figure 3 of the text) was examined after ion bombardment etching, cracks were seen in the cladding near the cladding-to-core bond layer. Some of these cracks can be seen in Figure A1. "Faxfilm" replicas were made and one of these replicas was prepared for electron microscopy. Another was examined with an optical microscope. Cracks in the cladding observed during the microscopic examination of the replica are shown in Figures A2 and A3. These cracks are very similar in appearance to the cracks shown in Figure A1. A part of the crack network shown in Figures A1. A part of the crack network shown in Figures A2 and A3 is shown in Figure A4 as it appeared in the electron microscope.

After the right face of piece No. 1 was etched, examined, and replicated, it was given a chemical etch with an HNO_3 -HF solution to reveal the grain structure of the cladding. Following this etch, the area shown in Figure A1 was reexamined and no cracks were seen. Figure A5 shows this area following the chemical etch. If the cracks seen in Figure A1 were real, they would be visible in Figure A5. It is believed therefore, that the cracks seen in the cladding following the etch by ion bombardment were a result of this etching method.

The foregoing discussion is included in this report as an interesting example of the artifacts which can occur during sample preparation.

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FIGURE A1

Cracks in Cladding Near Cladding-to-Core Bond Layer
(Etched by Ion Bombardment) 250X

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FIGURE A2

Cracks in Replica of Right Face of Piece No. 1
(Etched by Ion Bombardment) 250X

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FIGURE A3

Part of Figure A2 at Higher Magnification
500X

4-G1964B

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FIGURE A4

Electron Micrograph
of Part of Crack Network
Shown in Figures A2 and A3
2900X

FIGURE A5

Part of Area Shown in Figure A1
(Chemical Etch, Polarized Light)
500X

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