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FINAL REPORT: IRRADIATION PERFORMANCE OF COEXTRUDED
ENRICHED URANIUM FUEL ROD PT-IP-A172-A.

By T. T. Claudson

May 26, 1959

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**FINAL REPORT: IRRADIATION PERFORMANCE OF COEXTRUDED
ENRICHED URANIUM FUEL ROD PT-IP-A172-A.**

I. INTRODUCTION

The proposed operating conditions for fuel elements to be charged into the NPR require the fuel be of an extended surface geometry and maintain adequate strength and corrosion resistance in 300 C water. One fuel candidate for use in the NPR is the 7 Rod Cluster Fuel Element, the rods being fabricated by coextrusion of uranium with Zircaloy-2.

Contract No. AT(30-1) - 1565 was let to Nuclear Metals Inc. to produce by co-extrusion lengths of fuel rod containing both natural and enriched uranium of irradiation quality for fabrication into fuel elements. The fuel rods used in the irradiation test represent the first enriched uranium rods coextruded in 0.030 inches of Zircaloy-2 to be irradiated and examined at Hanford. The rods used for this test were fabricated into four, 4 rod cluster fuel elements thus allowing adequate space between individual rods for expansion in the case of a fuel rod failure. This rod was of particular interest since it contained an irregular uranium-Zircaloy-2 interface. The purpose of the irradiation was to determine the dimensional stability of coextruded fuel rods and to determine whether the irregularity in the bond interface had any effect upon the irradiation performance of the fuel. Goal exposure for the irradiation test was 2500 MWD/T.

II. SUMMARY

Four rod cluster fuel elements were irradiated in 200 C water in the KER Loop 2 facility to an exposure of 0.28 a/o burnup (2200 MWD/T). Post irradiation examination showed that each rod had increased an average of 0.008 inches in outside diameter and that microcracks had formed throughout the uranium core. The uranium had also increased in length to fill 0.050 inch of space left between the end cap and uranium for thermal expansion and uranium growth. A metallurgical bond between the end cap and the uranium had been formed during the irradiation. There was no effect of the irregular interface on the dimensional stability of the fuel rods.

Post irradiation examination included standard metallography, diametral and length measurements, replication, density and hardness measurements, and burnup analysis. The results of this irradiation test are described herein. No signs of ultimate fuel failure were observed. The coextruded 1.6% enriched fuel rod, irradiated in 200° C coolant with a calculated 510° C core temperature, operated successfully to an exposure of 2200 MWD/T.

III. FUEL FABRICATION HISTORY

A. Coextrusion of Rod

Uranium rods, 1.6% enriched, used in the irradiation were taken from extrusions Nos. 16781 and 16782 produced by Nuclear Metals Inc. The interface between the uranium and 0.030 inch Zircaloy-2 cladding was very irregular. A summary of the extrusion conditions is given in Table I. Table II lists the chemical and isotopic contents of the uranium billet used for the extrusion. (1)

(1) NMI 4710, Development and Properties of Coextruded Zircaloy - Clad Uranium Fuel Elements with Integral End Seals, H. F. Sawyer, Unclassified.

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TABLE I

Extrusion No.	Uranium Weight, grams	Nose Avg Composition % Cu	Preheat Temp. °F	Heating Media	Extrusion Cone (Included Angle) °	Die Diameter Inches	Extrusion Constant (tsi)	
							Start Run	
16781	19,490.2	100	1184	Horizontally in Graphite Crucibles	60	0.700	23.8	21.6
16782	19,492.5	100	1186		60	0.700	24.6	21.9

TABLE II

		Top	Bottom
Ingot No.	31026		
Isotopic Content	1.598±0.001%		
Density		18.93	18.94
Carbon		510	505
Nitrogen		15	15

ELEMENT	Al	B	Cd	Cr	Cu	Fe	Mg	Mn	Mo	Ni	P	Pd	SiO ₂	Sn	V	Zn
Top	8	0.2	0.2	10	5	33	6	8	4	10	20	1	120	1	20	20
Bottom	10	0.2	0.2	10	6	41	5	8	4	19	20	1	140	1	20	20

Analysis performed by National Lead Company of Ohio.
Constitutions given in parts per million.

The extrusion core of 1.6% enriched uranium was fabricated by National Lead Company of Ohio (Fernald Area) for delivery to NMI who then assembled the core into sealed extrusion billets. The core had been rolled to a 3-1/4 inch X 3-5/8 inch flat-faced oval, held in 1350 F carbonate salt for 30 minutes, water quenched, and finished machined to $2.583^{+0.000}_{-0.003}$ inches in diameter and 12 inches in length. This heat treatment was not adequate to refine the grain structure throughout the billet and subsequent extrusion resulted in an irregular uranium-zircaloy interface. The usual procedure for pre-worked stock at NMI is to beta heat treat at a diameter not more than 0.200 inch over the finished diameter. In subsequent extrusions this heat treatment error was corrected and proper interface conditions were achieved. Figure I is a photomicrograph showing the irregularity of the Zircaloy-uranium interface. The clad thickness varied between 0.025 and 0.032 inches.

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FIGURE 1

Photomicrograph of 1.6% uranium exposed to 0.28 a/o burnup at a calculated core temperature of 510 C. Irregular zr-2-uranium interface is evident. Cathodic etch. 1.5X.

B. Assembly of a Rod Cluster Fuel Element

Sections of rod from extrusion Nos. 16781 and 16782 were cut 8.500 inches in length and 0.250" of uranium bored out of each end to facilitate insertion of end caps. To insure that no uranium weld contamination could exist, the bored out ends of the rods were exposed to a hot nitric acid bath until all traces of uranium in the weld area were removed. End caps inserted into the ends were welded into place by the electron beam welding process. The rods were then beta heated in a trichloride salt bath at 710 C for five minutes and air cooled. Slots were machined into the end caps to accept a circular ring spider. The rods were then cleaned and pickled in HNO_3 -HF solution and nondestructively tested in an autoclave for 100 hours at 300 C. External pencil clip supports were then welded to both ends of each fuel rod and the assembly completed by welding four rods to the circular support. Five such elements were fabricated and four subsequently charged into the KER Loop 2 facility on June 30, 1958. Figure 2 shows the completed elements before irradiation.

The elements were irradiated in 200 C coolant at 1200 psi. The average specific power calculated from adjacent tube powers was 90 kw/ft. The calculated core temperature was 510 C. The test ran successfully without fuel failure to an exposure of 0.28 a/o burnup (2200 MWD/T). Figure 3 is a plot of the loop inlet temperature as a function of time in days, thus indicating the number of thermal cyclings to which the fuel was subjected during the irradiation.

IV. POST IRRADIATION EXAMINATION

After discharge, one fuel element was supplied to Radiometallurgy Laboratory Operation for examination. Figure 4 shows this fuel element after irradiation and prior to examination. The rods were separated and examined for visual signs of corrosion, bumping, hot spots, warpage, and other signs of impending fuel failure. No evidence of damage could be observed. A heavy iron oxide film existed on each rod over the heat generating surface. The fuel supports and end caps remained free of this film. This type of film has been observed before on similar fuel rods exposed to hot water which is acidic. Material exposed to hot water containing lithium hydroxide has not exhibited this film. The fuel rod diameter was measured and sections prepared for metallographic examination, density and hardness measurements, and burnup analysis. In each rod examined macrocracks and diameter increases of 0.005-0.012 inch were observed. To determine the extent of the damage incurred by the fuel rods, the remaining three elements were examined. In all sixteen rods, macrocracks and diameter increases up to 0.012 inch were observed.

From the nature of these cracks it was not immediately evident as to how they were formed. Metallographic specimens were prepared by standard polishing techniques with the final polish being with Linde A polishing compound. A cathodic bombardment technique was used in etching the specimens. Examination of the uranium core showed that in each rod the central two thirds of the rod had recrystallized in the alpha range. No evidence of beta transformation was evident. Macrocracks in the structure were most prevalent in the recrystallized portion of the rod. The cracks were random throughout the structure and not intergranular, but did in many cases run perpendicular to twin lines still evident in the structure. Both transverse and longitudinal sections were similar, indicating that the cracking was homogeneous in three dimensions.

FIGURE 2

As fabricated 4 rod cluster fuel elements before irradiation in the KER loop 2 facility.

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FIGURE 3

Effluent temperature vs. days in reactor illustrating coolant temperature variation during irradiation.

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FIGURE 4

Four rod cluster fuel element after 2200 MWD/T exposure
in 200 C coolant.

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Figure 5 shows a photomacrograph of a uranium fuel rod exposed to 0.28 a/o burnup illustrating the cracking in the central portion of the rod. Figure 6 illustrates the typical uranium structure found after irradiation in both the central area and near the edge of the rods. Lack of microstructure in the uranium is evident near the bond layer at this burnup and shows no evidence of recrystallization.

In only one rod was any damage to the bond observed. In this case, intergranular cracking was observed in the center of the rod. Three microcracks were found progressing along the uranium side of the bond and in one place through the bond layer. Photomicrographs are shown in Figure 7.

The fact that such cracking does not propagate along the bond itself demonstrates the strength and integrity of such a bond produced by coextrusion. Longitudinal sections of the end cap-uranium interface area were prepared and examined. Figure 8 shows this area before and after irradiation. The 0.025 inch space left for expansion and growth of the uranium has been completely filled and evidence of uranium creeping into the area between the end cap and cladding can be seen. Flow lines evident in the uranium structure show that the uranium had flowed from the hot, central part of the rod into the voided area. Such observations indicate that the length increase observed was the result of creep into the unrestrained region of the fuel rod.

Samples from the ends and center of one rod were prepared to measure the change in density during the irradiation. These samples were prepared by taking one half-inch wafers from the rods and machining off the clad. To insure that all clad was removed, the specimens were exposed to a hot nitric-hydrofluoric acid bath. Two other samples from the center of two rods were dissolved and submitted for chemical burnup analysis. The results of all post-irradiation measurements are given in Table II.

Density measurements indicated a major decrease in density which is a result of both swelling and the observed cracking. Density measurements given in Table II should be considered as the apparent density of the material since an accurate estimate of the crack volume is impossible. Replications for electron microscopy were prepared for study of the formation of swelling voids in the uranium structure. These studies were carried out by T. K. Bierlein and B. Mastel. Voids were found throughout the structure with diameters up to 240 Å. Generally speaking the larger swelling voids were seen in the center of the rod where the temperature was higher. Smaller voids occurred near the edge of the rod. Of particular interest were swelling voids which aligned themselves along twin and grain boundaries. Examples of this formation are seen in Figures 9 and 10.

It has been shown that variation in heat treatment alters the mechanical properties of the Zircaloy-2-uranium bond. Examination of the bond layer after irradiation was done to determine any change which might have occurred during the irradiation test. Replicas were prepared from unirradiated rod from the same extrusion for comparison. Electron microscopy studies revealed no evidence of any change during the irradiation. Examination of the replica taken from the irradiated rod did show what appeared to be fine microcracks in the clad. These cracks appeared to be 35-50 microns in length and

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FIGURE 5

Photomicrograph showing the cracking exhibited by 1.5% enriched uranium-zircaloy 2 clad rods after 2200 MWD/T exposure. Calculated core temperature was 510 C. 5X as polished.

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Photomicrograph showing typical uranium structure in central area of 1.6% enriched rods after 0.28 a/o burnup. 250X cathodic etch.

Photomicrograph showing typical uranium structure near edge of 1.6% enriched uranium rod after 0.028 a/o burnup. 250X cathodic etch.

FIGURE 6

Central core region inter-
granular cracking of 1.6%
enriched uranium exposed
to 0.28 a/o burnup. ➔
Cathodic etch. 250X.

Photomicrograph illustration cracking along and through zirconium-2 uranium bonding layer.
Material 1.6% enriched uranium exposed to 0.28 a/o burnup. Cathodic etch. 500X.

FIGURE 7

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A Before Irradiation

B After 0.28 a/o Burnup

FIGURE 8

Photomicrographs showing the filling of space left at the end cap for expansion of uranium.

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FIGURE 9

Negative print of a negative replica illustrating swelling void along twin boundaries. Material 1.6% enriched uranium exposed to 0.28 a/o burnup at a calculated core temperature of 510 C. 21,000X.

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FIGURE 10

Negative print of a negative replica illustrating swelling voids along grain boundaries. Material 1.6% enriched uranium exposed to 0.28 a/o burnup at a calculated core temperature of 510 C. 21,000X.

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TABLE II

Rod No.	Rod Diameter		Rod Length		Calc. ΔV	a/o Burnup	R	Density Grams/cc
	Position	Before	After	Before	After			
1-A-9	End	0.631	0.6410					
	Middle	0.631	0.6414	7.985	8.095	+4.6%	0.28	18.22
	End	0.631	0.632					
2-A-12	End	0.631	0.638					18.47
	Middle	0.631	0.638	8.000	8.110	+3.6%	0.28	18.17
	End	0.631	0.636					18.50
1-A-1	End	0.631	0.643					
	Middle	0.631	0.636	7.870	7.980	+5.3%	0.28	18.9
	End	0.631	0.640					
2-A-10	End	0.631	0.631					
	Middle	0.631	0.636	7.930	8.040	+2.8%	0.28	10.0
2-A-7	End	0.631	0.635					
	Middle	0.631	0.638	8.000	8.110	+3.6%	0.28	12.9
2-A-4	End	0.631	0.630					
	Middle	0.631	0.635	7.980	8.090	+2.4%	0.28	8.6
1-A-8	End	0.631	0.634					
	Middle	0.631	0.638	8.000	8.110	+3.6%	0.28	12.9

a very small fraction of this in width. An electron photomicrograph of this phenomenon is shown in Figure 11. From the maximum diameter increase measured, the strain in the clad was 1.90%.

As mentioned previously, it was found that the uranium had increased in length to fill the void left between the uranium and the end cap. Microscopic examination showed evidence that bonding of the central portion had occurred. Further examination by electron microscopy revealed distinct metallurgical bonding between the end cap and uranium similar to that produced between the clad and uranium by coextrusion. An electron photomicrograph of this is shown in Figure 12.

V. CONCLUSION

Sixteen 1.6% enriched uranium rods coextruded in 0.030 inches of Zircaloy-2 were irradiated to an exposure of 2200 MWD/T and examined. Each rod was found to have extensive macrocracking in the center of the rod and diametral increases up to 0.012 inch were found. No evidence was found which would indicate that the damage incurred would cause a rupture in the clad with

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FIGURE 11

Negative print of a negative replica showing possible clad cracking in rods exposed to 0.28 a/o burnup. Cathodic etch. 6000X.

FIGURE 12

Negative print of negative replica illustrating bonding which occurred in-reactor between the zircaloy-2 end cap and 1.6% enriched uranium coextruded rod exposed to 0.28 a/o burnup. Calculated core temperature 510 C. Cathodically etched. 11,000X.

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subsequent fuel failure. These cracks were probably the result of the tensile stresses imparted to the uranium core during the cooling cycle in the reactor when neutrons were not present in the fuel. Similar cracking has not been found in natural uranium rods coextruded in the same manner with Zircaloy 2 with exposures up to 1200 MWD/T. Such cracking phenomena are probably dependent upon the amount of exposure incurred by the specimen and the amount of enrichment present. Similar enriched material with exposures of 1250 MWD/T and 1600 MWD/T are awaiting examination and should reveal an exposure limit at which severe cracking begins. Examination of material now in the reactor with goal exposures of 5000 MWD/T should completely illustrate the irradiation capabilities of such fuel material. This test should be completed in December, 1959.

T. T. Claudson

T. T. Claudson
FUEL ELEMENT DESIGN OPERATION
HANFORD LABORATORIES OPERATION

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