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Report on Experiment
11f(1): "Investigation
of Barrier Layers for
Extruded Zircaloy-2 Clad
Molybdenum-Uranium Rods"

(PWR)

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Attached is the report on Experiment 11f(1): "Investigation of Barrier Layers for Extruded Zircaloy-2 Clad Molybdenum-Uranium Rods".

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INVESTIGATION OF BARRIER LAYERS FOR
EXTRUDED ZIRCALOY-2 CLAD MOLYBDENUM-URANIUM RODS

Introduction

This is a summary to date on the manufacture of 0.3" diameter rods consisting of a 12 w/o molybdenum-uranium fuel separated from its 30 mil clad by a thin layer of some other metal. This work was done under experiment 11f(1), "Investigation of Barrier Layers for Extruded Zircaloy-2 Clad Molybdenum-Uranium Rods", originally a part of the Process Development Section's work program. Recent corrosion data have shown that the corrosion resistance of a Zircaloy-2 clad 12 w/o molybdenum-uranium fuel rod decreases as the fuel-clad diffusion zone increases in thickness, the diffusion zone acting as a rapidly corrodible corridor. One method to eliminate the detrimental corrosion effects of the fuel-clad diffusion zone appeared to be the elimination of the Zircaloy-2 to molybdenum-uranium diffusion zone by separating the fuel and clad with another metal.

Procedure

After carefully washing all components as per established PWR practice, barrier layer elements were made by wrapping a foil of the barrier metal around a 12 w/o molybdenum-uranium fuel billet and then fitting this assembly into a Zircaloy-2 extrusion jacket and extruding. After extrusion, all rods were radiographed and sections were taken from the nose, middle and tail for metallographic examination. A great deal of the extruded rod was sent to Metallurgy for corrosion and other studies. Some stock was used for the manufacture of bond strength specimens. These latter specimens consisted of a rod about 3/4" long with the fuel and part of the Zircaloy-2 wall drilled out of each end to the extent that a thin flat wafer of fuel was left midway between the two ends. The pressure required to punch this wafer out, using a punch having a diameter slightly smaller

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than the fuel diameter, was assumed to be a measure of the fuel-clad bond strength (WAPD-FE-489). The disposition of all rod stock is shown in Table I.

A small effort was made to spray a barrier layer on a fuel billet prior to extrusion, using the Mogolelectric Metallizing Process and molybdenum powder and wire.

Results

All 12 w/o molybdenum-uranium fuel billets were from single uranium induction melted ingots except those used in extrusions 11f(1)-13 and 11f(1)-14, which were from stock that had been consumably melted after vacuum induction melting. A summary of the extrusion data is shown in Table I. The barrier layer did not completely wrap around the fuel in the first eight assemblies due to the skewing and upward movement of the foil during assembly caused by the close fit. Assemblies 11w(1)-1 and 11w(1)-7 were the worst in this respect and as a result a large portion of these rods did not show a barrier layer at all. For later assemblies the foil was overlapped at least a quarter to a half the fuel diameter and the dimensions were set such that a 3 to 4 mil clearance existed between the wrapped billet and clad component. Zircaloy-2 clad 12 w/o molybdenum-uranium rods with molybdenum, tantalum, sponge zirconium and niobium layers having final thicknesses as shown in Table I, were extruded. It can be seen from this table that the total press capacity was necessary to extrude to a 25:1 reduction. In one case, the extrusion was incomplete and in another, the extrusion was completely unsuccessful.

Figures 1 through 8 show three of four different barrier layers at various magnifications. For all four barriers, metallographic examination revealed intimate fuel and clad contact with the barrier, although in no case was there any diffusion evident at magnifications as high as 370X. The niobium barrier had a dimensional

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appearance quite similar to that of the sponge zirconium barrier, pictured in Figure 8. It is apparent by a comparison of Figures 1 and 2 with Figures 3 and 4 that the tantalum barrier is smoother than the molybdenum barrier and appears to have suppressed to a great extent the microscopic core-clad variations common to many bi-metal extrusions. Limited evidence, however, as shown in Table IV, does appear to indicate that rods extruded with a 0.3 mil layer of molybdenum, or a 2 mil layer of niobium, result in better macroscopic fuel dimensions than those with a 1 mil layer of sponge zirconium. Figure 7 and Table III show the great thickness variation of the 0.3 mil molybdenum barrier. It is apparent from the table that the percentage variation in thickness decreases as the barrier thickness increases for molybdenum and also that the tantalum barrier is more uniform than the molybdenum barrier of equal thickness.

Figures 5 and 6 show the incomplete junction of the metal foil that occurred in the first eight extrusions.

The fuel-clad strength for several different barrier conditions is shown in Table II and it appears that the bond strength increases as the barrier thickness decreases.

The need for different thicknesses of barrier layers required that foil be made to thicknesses not immediately available. This was done by cold rolling larger stock. An as-received 5 mil sheet of molybdenum was successfully cold rolled, without annealing, between two 0.75" thick heli-arc welded Ti-Namel plates to 1.5 mils. The Ti-Namel took a 5.5:1 reduction to reduce the molybdenum 3.3:1. Niobium was also cold rolled without edge cracking from 5 mils to 1 mil in the same manner.

An attempt was made to spray molybdenum on 12 w/o molybdenum-uranium billets using 80 mesh molybdenum powder and the Mogolelectric Metallizing Process. The molybdenum appeared to oxidize quite rapidly in the air and formed a layer on

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the billet, the greater part of which spalled off during cooling. Next, a 1/16" diameter molybdenum wire was used and the feed of the metallizer was slowed down to a point where efficient melting of the wire occurred and a very strongly adherent film was sprayed on a scrap Zircaloy-2 fuel plate. Metallographic examination revealed that the molybdenum film was layered and discontinuous. This type of film was considered unsatisfactory and hence, no further work was done with this process.

Conclusions and Recommendations

Zircaloy-2 clad 12 w/o molybdenum-uranium rods having molybdenum, tantalum, sponge zirconium and niobium barrier layers between the clad and fuel with final thicknesses of from 0.3 to 2 mils, have been extruded from a 1.6" container to 13:1 and 25:1 reductions. Except for the molybdenum layer, all of the barrier layers appear to be reasonably uniform in thickness. All show intimate contact with the fuel and clad but with no apparent diffusion, as observed at 370X magnification.

A number of rods have been transferred to Metallurgy for corrosion testing, the results of which will be reported in their progress reports and the minutes of their meetings with the Uranium Alloy Panel.

Continued interest has been shown in a barrier layer type element and work is in process to manufacture elements containing platinum, palladium, molybdenum and niobium metal and alloy barriers.

Since the press capacity was required to extrude the Zircaloy-2 clad 12 w/o molybdenum-uranium billets reported herein to a 25:1 reduction from a 1.6" container, sometimes without success, it is suggested that no extrusions of a service or production nature be made unless there is a reasonable probability of success.

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TABLE I

Summary of Extrusion Data

Extrusion No.	Fuel Ingot No.	Fuel Hardness Rc	Billet Temp. °F	Die Size in.	Reduction Ratio	Extrusion Rate in./sec.	Maximum Tonnage	Running Tonnage	Barrier	Barrier Thickness mils	Remarks and Disposition of Usable Material
11w1-1	Y270-1	34	1950	.426	13:1	0.5	160	140	Mo	2.8	29" is available.
11w1-2	Y270-2	28	1950	.437	13:1	0.5	145	145	Mo	2.8	27" is available.
11w1-3	Y271-1	29	1950	.314	25:1	0.5	170	150	Mo	2.0	50" is available.
11w1-4	Y271-2	30	1950	.314	25:1	0.5	Max.	160	Mo	2.0	46" transferred to Metallurgy, 4" used in bond test.
11w1-5	Y271-3	32	1950	.314	25:1	0.5	Max.	160	Ta	2.0	50" is available.
11w1-6	Y272-1	30	1950	.314	25:1	0.5	170	160	Ta	2.0	46" transferred to Metallurgy, 4" used in bond test.
11w1-7	Y272-2	32	1950	.426	13:1	0.5	150	130	Ta	2.8	30" is available.
11w1-8	Y272-3	33	1950	.437	13:1	0.5	160	140	Ta	2.8	26" is available.
11w1-9	Y268-1	29	1950	.314	25:1	0.1	Max.	Max.	Mo	0.3	30" extruded. 12" is available, remainder scrapped.
11w1-10	Y269-1	30	1950	.314	25:1	0.5	Max.	170	Mo	0.3	44" transferred to Metallurgy, 4" used in bond test.
11w1-11	Z16-2	26	1950	.314	25:1	0.5	Max.	160	Zr	1.0	48" transferred to Metallurgy.
11w1-12	Z18-2	31	1950	.314	25:1	0.4	Max.	160	Zr	1.0	46" transferred to Metallurgy, 4" used in bond test.
11f1-13	J704-3	32	1935	.326	25:1	1.0	Max.	Max.	Nb	2.0	50" is available.
11f1-14	J666-2	31	1930	.326	25:1	0.8	Max.	Max.	Nb	---	Did not extrude. Material scrapped.

- NOTE:
1. Billet preheat - 9 minutes.
 2. Heating medium - Liquid Heat 1550 - 85% BaCl₂, 15% NaCl.
 3. Back-up plug - copper.
 4. Container size - 1.6"
 5. Container temperature - 750°F
 6. Die temperature - 700°F
 7. Lubrication - Led Plate
 8. Press capacity - 180 tons

TABLE II
Fuel-Clad Bond Strength

<u>Extrusion No.</u>	<u>Barrier Layer</u>	<u>Avg. Load (psi)</u>	<u>One Sigma (psi)</u>	<u>No. Specimens Tested</u>
RA	None	46,566	2,710	3
11w1-4	2.0 mil Mo	37,225	3,313	4
11w1-10	0.3 mil Mo	47,625	3,047	4
11w1-6	2.0 mil Ta	30,300	7,532	3
11w1-12	1.0 mil Sp. Zr	52,775	6,749	4

TABLE III
Measurement of 0.3 Mil Molybdenum Barrier

<u>Extrusion No.</u>	<u>Barrier Layer</u>	<u>Avg. Thickness (mils)</u>	<u>One Sigma (mils)</u>	<u>No. Readings</u>
11w1-9	Mo	0.32	0.12	11
11w1-9	Mo	0.32	0.15	10
11w1-10	Mo	0.35	0.14	10
11w1-3	Mo	2.40	0.17	5
11w1-4	Mo	2.16	0.21	5
11w1-6	Ta	2.06	0.13	5

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TABLE IV

Dimensional Measurements of Barrier Layer Extrusions

Specimen No.	Barrier Layer	Outside Diameter			Fuel Diameter			Clad Thickness		
		Avg. Dia. (mils)	Std. Dev. (mils)	No. Readings	Avg. Dia. (mils)	Std. Dev. (mils)	No. Readings	Avg. Dia. (mils)	Std. Dev. (mils)	No. Readings
11w1-9	0.3 mil Mo	331.8	2.8	4	243.7	1.8	4	35.1	1.4	8
11w1-10	0.3 mil Mo	314.8	2.7	4	242.9	1.2	4	35.9	3.2	8
11w1-11	1.0 mil Zr	306.1	2.1	4	234.4	3.2	4	35.8	1.4	8
11w1-12	1.0 mil Zr	304.2	2.0	4	231.4	2.7	4	36.4	3.0	8
11f1-13	2.0 mil Nb	308.4	3.4	4	242.9	1.3	4	32.8	2.3	8

Zircaloy-2

2 Mil Tantalum

12 w/o Molybdenum-Uranium

Neg. No. 8479A-3

500 X

Figure 1

12 w/o Molybdenum-Uranium Fuel Separated From
Zircaloy-2 Clad by a 2 Mil Tantalum Layer

Zircaloy-2

2 Mil Tantalum

12 w/o Molybdenum-Uranium

Neg. No. 8479A-1

100 X

Figure 2

12 w/o Molybdenum-Uranium Fuel Separated From
Zircaloy-2 Clad by a 2 Mil Tantalum Layer

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Zircaloy-2

2 Mil Molybdenum

12 w/o Molybdenum-Uranium

Neg. No. 8477A-3

500 X

Figure 3

12 w/o Molybdenum-Uranium Fuel Separated From
Zircaloy-2 Clad by a 2 Mil Molybdenum Layer

Zircaloy-2

2 Mil Molybdenum

12 w/o Molybdenum-Uranium

Neg. No. 8477A-4

100 X

Figure 4

12 w/o Molybdenum-Uranium Fuel Separated From
Zircaloy-2 Clad by a 2 Mil Molybdenum Layer

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Zircaloy-2

2 Mil Tantalum

12 w/o Molybdenum-Uranium

Neg. No. 8479A-2

250 X

Figure 5

Incomplete Junction of 2 Mil Tantalum Barrier

Zircaloy-2

2 Mil Molybdenum

12 w/o Molybdenum-Uranium

Neg. No. 8477A-1

250 X

Figure 6

Incomplete Junction of 2 Mil Molybdenum Barrier

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Neg. No. 8732A-1

500 X

Figure 7

12 w/o Molybdenum-Uranium Fuel Separated From
Zircaloy-2 Clad by 0.3 Mil Molybdenum Layer

Neg. No. 8734A-1

100 X

Figure 8

12 w/o Molybdenum-Uranium Fuel Separated From
Zircaloy-2 Clad by 1 Mil Sponge Zirconium Layer

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