NOTICE

**CERTAIN DATA
CONTAINED IN THIS
DOCUMENT MAY BE
DIFFICULT TO READ
IN MICROFICHE
PRODUCTS.**

Received

FEB 08 1991

DISTRIBUTION

F. E. Kruesi, Wilm.
J. E. Conaway
T. Hendrix, SRP
H. J. Groh
J. L. Womack
J. Jiunnies
H. E. Wingo
G. F. Merz
S. P. Rideout
M. K. Carlson

B. S. Spangler
F. D. Benton
L. R. Chandler
D. B. Rankin
S. Mirshak, SRL
J. M. Boswell
M. M. Anderson
D. A. Sharp
R. G. Willard
J. A. Porter

M. L. Hyder
D. R. Johnson
T. J. Anderson
R. S. Ondrejcin
J. P. Morin
W. M. Massey
D. R. Muhlbaier
H. P. Olson
R. L. Frontroth
TIS File (2)

August 20, 1980

MEMORANDUM

TO: D. A. WARD

FROM: H. B. PEACOCK - E. F. STURCKEN

INSPECTION OF IRRADIATED P-7 FUEL TUBES

INTRODUCTION

Mark 16 U-Al alloy production fuel tubes and six special U_3O_8 -Al powder metallurgy (PM) test assemblies⁽¹⁾ were successfully irradiated in P-7 reactor charge beginning December 1976. A year after irradiation, the outer surfaces were inspected under water in P-Area basin. Inspection showed that a "black" oxide had formed on the bottom 2/3 and flaked off in some areas for both the production and PM tubes. A small cladding defect was also observed on one PM outer tube near the bottom.

Sections were cut from the tubes and metallographically examined in the SRL High Level Caves (HLC). This report gives results of the examinations.

SUMMARY

Both U-Al alloy and U_3O_8 -Al tubes had "black" oxide on the bottom 2/3 of the tubes. The oxide thickness ranged from about 1/2-1 mil at the top to 2-1/2 mils thick near the bottom tube end where the oxide flaked off. X-ray studies showed that the "black" oxide was $Al_2O_3 \cdot H_2O$ (Bohmite) a high temperature form of alumina. "Black" oxide has been produced on 8001 aluminum in the TNX corrosion test loop when the heat flux was 10^6 pcu/hr and the coolant temperature $>75^\circ C$.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

A hole observed on one U_3O_8 -Al tube resulted from basin corrosion. The core was not attacked, only the outer cladding. Pitting corrosion was also observed near the hole on the inner cladding. No cladding thinning, oxide segregation, or inclusions were found in the core. Diffusion controlled U_3O_8 -Al reactions did not thin the cladding of the highly loaded core.

Fuel Tubes and Irradiation

Production tubes with U-Al alloy cores contained about 28 wt % uranium while the PM tubes had 47 wt % U (56% U_3O_8). The PM tubes were made by the PM process being developed by SRL.⁽²⁾ All tubes were extruded in 321-M.

The experimental assemblies were successfully irradiated in Gang I of P Reactor to a fission density of $\sim 1.6 \times 10^{21}$ fissions/cm³. The average operating power determined from RDAP's for the PM and production assemblies was 7.3 and 6.5 mw, respectively. The average effluent temperature was 102°C and the maximum heat flux was 0.43×10^6 pcu/hr-ft². The nominal heat flux for a Mark 16 assembly is 0.33×10^6 for a 6.5 mw assembly.⁽³⁾

The performance of fuel tubes in the reactor is qualitatively evaluated by measuring the ^{92}Sr activity of the moderator. Small leaks of ~ 100 mg uranium can be detected. The ^{92}Sr activity for P-7.1 to 7.4 subcycles is shown in Figure 1. The activity remained practically constant or decreased slightly near the end of the cycle. No cladding failures were expected. The decrease in activity in March resulted from a shutdown caused by malfunctions of reactor instrumentation, during which $Gd(NO)_3$ solution was injected into the moderator.

Typical photomicrographs of unirradiated and irradiated U-Al alloy and U_3O_8 -Al cores are shown in Figures 2 and 3, respectively. The microstructure of the U-Al alloy before and after irradiation appears similar. The aluminides (UAl_x) are uniformly dispersed in the matrix. The black spots in the irradiated microstructure are probably irradiation-induced voids. However, small graphite inclusions were also observed.

The U_3O_8 -Al microstructure is significantly different after irradiation. The as-fabricated microstructure shown in Figure 2a is a uniform dispersion of U_3O_8 in an aluminum matrix. During irradiation a diffusion controlled reaction occurs forming UAl_x type particles. The voids in the PM fuel particles occur from: 1) less than theoretically dense U_3O_8 particles, 2) fabrication, and 3) U_3O_8 -Al reactions. Voids are sites for fission gases which reduces the propensity for swelling.

Basin Examination

Outer surfaces of the fuel assemblies were inspected (see Figure 4) in P-Area basin⁽⁴⁾ 1 year after discharge from the reactor. Both production U-Al alloy and U_3O_8 -Al tubes had a "black" oxide on the lower 2/3's of the tubes. Areas were also observed near the bottom where the oxide had flaked off. A hole was found on one U_3O_8 -Al outer tube at the oxide interface. Photographs taken through the periscope in the basin are shown in Figure 5.

Six- and ten-inch-long sections were cut from the top and bottom, respectively, of one each U-Al and U_3O_8 -Al tube for further examination in the HLC. Locations of the sections are given in Table I.

HLC examinations

The as-received samples are shown in Figure 6. Specimens from the top of both tubes exhibited a "white" oxide surface appearance while those from the bottom looked "black" and had areas where the oxide had flaked off. These areas are shown in the above figure.

Half inch samples were cut and mounted for metallographic examination to determine the oxide thickness and the extent of core reactions. Figure 7 shows the oxide thickness for U-Al and U_3O_8 -Al tubes. The oxide thickness was similar regardless of the type core. For the top section, the "white" oxide layer was 1/2-1 mil thick while the "black" oxide at the bottom was about 2.5 mils thick. Oak Ridge measured the oxide thickness for aluminum and found that above 2 mils the oxide flakes off.⁽⁵⁾ This is consistent with the fact that the oxide flaked off near the bottom where it was greater than 2 mils. The thick oxide layers showed many cracks parallel and perpendicular to the tube surface. About 0.5 mil of aluminum is needed for every mil of aluminum oxide.

A "black" oxide has been observed on Mark 16 fuel tubes in C-1 (1971)^(6,7) and assumed to be caused by "dogboning" which increased the operating temperature in the area. Near the bottom of these tubes were cladding penetrations which appeared to result from thin cladding. Pits were also present along longitudinal scratches.

The diffusion controlled U_3O_8 -Al reactions at the cladding-core interface did not thin the cladding. From the photographs, the cladding thickness ranged from 33 to 35 mils for the U-Al alloy production tube and 35-39 for the PM tube. The reaction produces a uranium-aluminum compound ($U-Al_x$) which has not been identified by x-ray diffraction because of irradiation damage.

A 1-inch-square section was cut from the PM tube which included the 1/2-inch-diameter hole. The section was mounted in the transverse direction and ground in 0.030- to 0.050-inch increments. Photographs were made after each grinding. No evidence of cladding thinning or powder segregation/agglomeration was found. A photograph made near the center of the hole is shown in Figure 8a. Examination showed only cladding corrosion. The core was not attacked. After polishing for metallographic analysis, a corrosion pit was observed on the inner cladding as shown in Figure 8b.

Corrosion occurred in the basin because of the following facts: 1) all the core was present (no core undercut as observed for cladding failures in the reactor), 2) presence of corrosion products (powdery oxide) around the hole, and 3) formation of corrosion pits in areas where the oxide flaked off. The corrosion products would have been washed away had the failure occurred in the reactor.

X-ray Diffraction Studies of "Black" and "White" Oxides

Powder samples were scraped (in the High Level Caves) from the "black" region at the bottom and the "white" region at the top of the tube for both Mark 16 U_3O_8 -Al and U-Al fuel tubes. X-ray diffraction patterns were obtained with a Debye-Scherrer camera and confirmed with x-ray diffractometer traces.

The "black" oxide was $\alpha\text{Al}_2\text{O}_3 \cdot \text{H}_2\text{O}$, Böhmite a high temperature form of alumina and the "white" oxide was $\alpha\text{Al}_2\text{O}_3 \cdot 3\text{H}_2\text{O}$, Gibbsite. Gibbsite is the usual form found in the reactor. "Black" oxide has been formed on 8001 under simulated reactor conditions by R. S. Ondrejcin⁽⁸⁾ in the TNX loop when the cooling water temperature was raised from 50 to 75°C and the heat flux to 10^6 pcu/h. The high temperature oxide was Böhmite, as observed in the present case.

Scanning Electron Microscopy (SEM) and X-ray Energy Spectroscopy (XES)
Studies of "Black" and "White" Oxides

During sampling, it was observed that the "black" oxide was scaly and more crystalline in appearance than the "white" oxide which was more powdered in appearance. The SEM micrographs, Figure 9, confirm these observations.

XES spectra of the "white" oxide showed only alumina; however, the "black" oxide contained impurities of silicon, chlorine, sulfur, calcium, and iron, Figure 9. These impurities were probably picked up in the storage basin since the "black" oxide was in contact with corroded steel rollers when it was raised in the storage basin for observation.

Future Work

The formation of "black" oxide is routinely observed on tubes from all reactor charges. It is recognized that increases in heat flux or coolant temperature may cause the oxide to exceed its critical thickness and spall from the surface. Projected reactor power improvements may affect the oxide behavior. It is therefore recommended that an out-of-reactor SRL program be initiated to determine the maximum operating limits for 8001 aluminum alloy. Also, other high temperature aluminum alloys should be evaluated for use in SRP reactors.

HBP/EFS:ehj

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

REFERENCES

1. Peacock, H. B., Quality of U_3O_8 -Al Fuel Tubes for the P-7 Irradiation Test, DPST-77-247, February 1977.
2. SRL Monthly Report, "Development of U_3O_8 -Al Powder Metallurgy Fuel," DP-75-1-8, August 1975.
3. Hydraulic and Heat Transfer of Mark 16 and Mark 16-B Assemblies, DPSTM-16(H), November 1976.
4. SRL Monthly Report, "Quality of Irradiated U_3O_8 -Al Fuel Tubes," DP-78-1-12, December 1978.
5. Oak Ridge Reactor Chemistry Progress Report, "Effect of Heat Flux on Corrosion of Aluminum by Water," ORNL 3262, pp 92-96.
6. Angerman, C. L., Cladding Penetrations in Irradiated Mark 16 Elements, DPST-71-506, September 23, 1971.
7. Alewine, G. B. and R. S. Wingard, Mark 16 Fuel Assemblies, DPST-71-304, April 13, 1971.
8. Ondrejcin, R. S., Corrosion of 8001 Aluminum Fuel Cladding, DPST-72-371, May 31, 1972.

TABLE I
OUTER SPECIMENS FOR HLC EXAMINATION

	<u>U₃O₈-A1</u>	<u>U-A1</u>
Tube number	0 x 1010	-
X-Y coordinates	24 x 30	28 x 36
Wt % uranium	47	~28
Core length, inches	154	154
Center of sample * to bottom of core	102-3/4 39-1/4	102-3/4 39-1/4

* From bottom of core as positioned in the reactor.

FIGURE 1. END OF MONTH ^{92}Sr MODERATOR ACTIVITY FOR P-7.1 TO 7.4 SUBCYCLES

a. Unirradiated

100X

b. Irradiated

100X

FIGURE 2. PHOTOMICROGRAPHS OF UNIRRADIATED AND IRRADIATED U-AL ALLOY
CORES (as-polished)

a. Unirradiated

100X

b. Irradiated

100X

FIGURE 3. PHOTOMICROGRAPHS OF UNIRRADIATED AND IRRADIATED U_3O_8 -Al
CORES (as-polished)

FIGURE 4 . BUNDLED U₃O₈-A1 FUEL TUBES IN P-AREA BASIN

a. Oxide - No Oxide Interface

5X

b. No Oxide Area

~7X

FIGURE 5 . OUTER SURFACE OF MARK 16 PM FUEL TUBES IRRADIATED IN P-7.1-7.4
(Photographed through periscope in P-Area basin)

a. Top Sections

b. Bottom Sections

FIGURE 6. SECTIONS OF OUTER FUEL TUBES SENT TO HLC FOR METALLOGRAPHIC EXAMINATION

FIGURE 7. OXIDE LAYERS FOR MARK 16 OUTERS WITH U-Al AND U₃O₈-Al CORES FROM THE TOP (WHITE OXIDE) AND BOTTOM (BLACK OXIDE) OF THE TUBES 46X

a. Center of defect

4.4X

b. Inner cladding

50X

FIGURE 8. SECTION THROUGH THE DEFECT ON THE MARK 16 PM OUTER FUEL TUBE

TOP
white oxide

BOTTOM
black oxide

FIGURE 9. SEM PHOTOMICROGRAPHS AND XES SPECTRA OF OXIDES FROM TOP (WHITE) AND BOTTOM (BLACK) OF TUBES

END

DATE FILMED

02 / 22 / 91