POSTIRRADIATION EXAMINATION OF TANTALUM CLADDING
FROM PHOTONEUTRON STARTUP SOURCES FOR EBR-II

by

R. V. Strain, D. C. Cutforth, and V. G. Eschen

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ABSTRACT

The tantalum cladding from EBR-II antimony-beryllium photoneutron startup sources was examined for evidence of corrosion and changes in physical properties. These sources were exposed to a 4-gpm flow of the EBR-II primary-system sodium during their residence in the reactor. A tantalum-clad source irradiated to a total peak fluence of about 3.4×10^{22} nvt was examined most thoroughly. Optical metallographic examination revealed minor pitting at the outside surface of the cladding but no evidence of serious corrosion or other microstructural changes. Chemical analysis indicated an increase of 709 ppm oxygen, 98 ppm nitrogen, and 11 ppm hydrogen in the irradiated cladding. Changes in density of 2% (maximum) and changes in microhardness as great as 230 DPH were also observed.

I. INTRODUCTION

Five tantalum-clad, antimony metal cylinders were fabricated for use in an antimony-beryllium photoneutron startup source in the Experimental Breeder Reactor II (EBR-II). The sources were fabricated by welding a lower-end fitting to a tantalum tube, 0.793 ± 0.001 -in. ID, 0.915 ± 0.001 -in. OD, and 14.75-in. long. A cylindrical antimony slug, 14-in. long with a diameter of 0.780 in., was placed in the tube and an upper-end fitting was welded on. The source (the tantalum-clad cylinder) is attached to a positioning fixture as shown in Fig. 1. The cylinder is positioned in a stainless-steel-clad beryllium sleeve, as shown in Fig. 2, to complete the source.

STAINLESS-STEEL POSITIONING FIXTURE

Fig. 1. Tantalum-clad Cylinder and Positioning Fixture

Fig. 2. Tantalum-clad Cylinder Inserted in Beryllium Sleeve to Form the Assembled Photoneutron Startup Source

The antimony cylinder was clad to provide mechanical protection for the relatively brittle antimony and to eliminate interaction between the antimony and the primary-system sodium coolant of the EBR-II. Tantalum was used for the cladding rather than the more conventional stainless steel to avoid potential problems in compatibility between stainless steel and antimony. Investigations at the Southern Research Institute¹ and at Battelle Memorial Institute² showed that, of the many materials tested, tantalum had the best compatibility with molten antimony and that stainless steel was particularly susceptible to attack from molten antimony. In-reactor irradiation temperatures above the melting point of antimony were not expected for the source, but tantalum was used instead of stainless steel in the interest of safety.

Four tantalum-clad antimony cylinders (referred to as "sources" hereafter in this report) were installed in the reactor grid during the startup of EBR-II. Three of them, SO-1911, SO-1912, and SO-1915, have been removed at intervals from the reactor to examine the tantalum cladding and evaluate its performance in reactor service. These examinations consisted of measurements of diameter and wall thickness and optical metallographic examinations.

Interest in using tantalum as control material in future LMFBR's resulted in a more comprehensive examination of SO-1915, the source most recently removed from the reactor. Postirradiation examinations of SO-1915 consisted of measurements of diameter and wall thickness; optical metallography; density determinations; and chemical analysis for oxygen, nitrogen, and hydrogen. A brief irradiation history for each of these sources during their residence in the EBR-II grid is given in Table I. The sources were in the blanket region of the reactor, where the neutron flux is not well characterized; consequently, the total fluences listed in Table I have an uncertainty of about $\pm 20\%$. When the antimony cylinder is not being used in the source, it is stored in the reactor core in a hexagonal stainless-steel cylinder similar in dimensions to the standard subassembly. Figure 2 shows that the midlength of the source was at the axial center line of the core during its residence in the reactor. The change in relative fluence in row 8 of the reactor with distance from the bottom of the core, as obtained from axial gamma scanning of an irradiated rod from a depleted-uranium blanket subassembly, is shown in Fig. 3.

TABLE I. Operating History of the Sources

Source Number	Grid Position	Insertion Date ^a	Reactor Power, MWd	Total Fluence, 10 ²¹ nvt
SO-1911	14B5	5- 9-63	Critical Testing ^b ↓	None ↓
	13B6	9- 4-63		
	14B5	9- 2-63		
	8E2	9-28-63		
	13B6	10-28-63		
	8F2	10-29-63		
	13B6	11-11-63		
	7E5	11-13-63		
	13B6	12- 3-63		
	16E9	6-23-64		
	11E1	6-24-64		
	13D6	6-24-64		
	Out	3-27-67	Total 12,247	Total 1.4
SO-1912	14B5	9-11-63	Critical Testing ↓	None ↓
	7E3	9-12-63		
	13E6	10-24-63		
	7E5	11-11-63		
	7E6	12- 3-63		
	13F6	6-23-64		
	8A4	3-24-66	Total 12,247	Total 10.6
SO-1915	7E2	6-23-64	9,880	9.0
	8E5	1- 9-67	26,274	21.0
	8E7	6-27-69	2,867	4.0
	Out	9- 5-69	Total 39,021	Total 34.0

a. The insertion date is also the date of removal from the previous position.

b. The reactor was operated at zero power until Aug. 1, 1964.

Fig. 3. Relative Axial Fluence Characteristics in Row 8 of the Reactor
(Obtained from an Axial Gamma Scan of an Irradiated Rod from a
Depleted-uranium Blanket Subassembly)

The irradiation temperatures of the sources were assumed to be similar to those of the adjacent subassemblies in the reactor grid because the sodium flow through the source assemblies is only about 4 gpm. Calculations indicated that the temperature of subassemblies in row 8 rises linearly from 700°F at the bottom of the core to 900°F at the top. Gamma heating from the source is not considered to contribute significantly to the temperature of the source assembly. Therefore, the irradiation temperature of source SO-1915 is estimated to have varied from 700°F at the bottom to 900°F at the top.

II. POSTIRRADIATION-EXAMINATION PROCEDURES

A. Sodium Removal

Normally, irradiated components transferred from the reactor core to the air cell of the FEF are subjected to a sodium-removal operation in which the residual sodium on the surfaces of the components is oxidized by moist air or argon and the surfaces are then rinsed clean with water. Because tantalum has poor resistance to corrosion by alkaline solutions, the sources were subjected only to a purge of dry air at 10 cfm for 2 hr to prevent contamination of the interbuilding coffin and to avoid corrosion of the cladding itself. This purge was to oxidize the outer surfaces of the residual sodium to reduce the fire hazard during subsequent handling of the source in the air cell. The source was transferred into the air cell, removed from its stainless-steel storage thimble, and transferred into the FEF argon-atmosphere hot cell. The argon-cell atmosphere is maintained at a nominal purity of 40 ppm O₂, 40 ppm H₂O, and 7% N₂. Total exposure time of the source to the air atmosphere, including the oxidation step, was less than 4 hr.

B. Sectioning and Sample Preparation

Source SO-1915 was sectioned by using a mechanical hacksaw in the argon cell. The samples from SO-1911 and SO-1912 were sectioned in the air cell. Three samples about 1/2-in. wide were taken, as shown in Fig. 4. Attempts to remove the antimony slugs from the tantalum cladding by tapping the sample against a mandrel with the master slave manipulators failed to

dislodge the antimony. The slugs were removed by slitting the tantalum cladding with a cutoff wheel. Because the antimony was not bonded to the tantalum, the slugs were then easily removed from the cladding. The 1/2-in. sections of the sources were transferred to the ANL-Idaho junior cave facility, which contains the analytical-chemistry and metallographic-preparation cells. Small samples of the cladding (1/8 x 1/4 x 1/16 in. for metallography and 3/8 x 1/4 x 1/16 in. for chemical analysis) were cut from the samples. The antimony slugs were returned to the FEF for disposal.

The high activity of the samples presented a problem during sectioning and examination in the cave facilities. The sections containing the antimony slugs caused radiation levels of 80 mr/hr on the operator side of the 2-ft-thick, high-density concrete shielding of the cave hot cells. The metallographic and chemistry samples were cut by using a water-cooled, abrasive cutoff wheel in a closed box within the cell. The oxidation of tantalum produces a light, fluffy oxide powder that is easily transported by air currents. The nine cuts required to obtain the metallographic samples produced enough of this powder to increase the radiation level in the absolute filters of the ventilation system for the metallographic-preparation cell from 100 mr/hr to ~25 R/hr at 1-ft distance. The water used during the cutting exhibited about 100 R/hr at the same distance. These readings were taken shortly after the source was cut, but about 45 days after the source was removed from the grid. The tantalum sample used for chemical analysis had an activity of 25 R/hr at 1 ft 75 days after it was removed from the reactor. The metallographic samples produced a field of about 100 mr/hr through the 6-in.-thick steel shielding of the metallographic-examination cell.

Future examinations are expected to be carried out in the Hot Fuels and Examination Facility, where adequate shielding will be available.

C. Chemical Analysis

Oxygen, nitrogen, and hydrogen were determined by using an inert-gas, fusion technique. The samples were melted in a graphite crucible and the gases were picked up in a stream of helium, which was passed over copper

oxide. Reaction with the graphite converted the oxygen to carbon dioxide and reaction with the copper oxide converted the hydrogen to water. The gases were then separated by using cold traps. The water was condensed in an ice bath and the carbon dioxide in a liquid-nitrogen bath, with the nitrogen being collected on molecular sieves at liquid-nitrogen temperatures.

D. Metallographic Examination

The metallographic samples were mounted in a cold-setting epoxy resin (Epon 812) and ground by using 120-, 320-, 400-, and 600-grit silicon carbide paper. The samples were polished with 6- μ diamond paste, then 1- μ and 0.3- μ aluminum oxide on vibratory polishers. The samples were etched with HNO_3 , HF, and H_2O in equal proportions. The microstructure of the samples was brought out by alternately polishing and etching them several times.

E. Density Determinations

The samples were prepared for the density determinations by pickling in nitric acid, and then rinsing, first in deionized water then in alcohol. The densities were determined by the immersion (Archimedes) technique. A constant-temperature bath was used to maintain the carbon tetrachloride immersion media at a constant temperature of 25.2°C. The densities of the irradiated samples and unirradiated control samples were measured with the same apparatus.

F. Microhardness Measurements

Microhardness was measured with a remotely operated "Tukon" microhardness tester. The accuracy of the unit checked within 1% of a certified standard having a hardness of 198 (standardized using a 500-g load) at the start and finish of the measurements.

G. Diameter Measurements

Diameters of sources SO-1911 and SO-1912 were measured with a hand micrometer. The measurements for SO-1915 were made with a dial indicator mounted in a special fixture. A machined rod 1 in. in diameter was used as a standard.

III. RESULTS AND DISCUSSION

A. Chemical Analysis

The results of chemical analyses of a sample of the cladding from near the midlength of SO-1915 and a control sample are given in Table II. These results show an appreciable increase in the oxygen, nitrogen, and hydrogen content of the cladding. Because of the long time that the sources were in the reactor, these values should be the equilibrium values for these elements in the cladding in contact with the EBR-II primary sodium.

TABLE II. Chemical Analyses of Tantalum Cladding
(ppm by weight)

Source Number	Oxygen	Nitrogen	Hydrogen
Unirradiated	94	9	2
SO-1915	803	107	13

B. Metallographic Examination

Photomicrographs of typical areas of the microstructure of samples from the unirradiated tantalum and from SO-1915 are shown in Figs. 5 and 6. The optical metallographic examination of samples from the sources revealed some evidence of minor pitting of the outer surface of SO-1915. The pits are assumed to have occurred while the source was in the reactor

Fig. 5. Photomicrographs of Typical Areas of the Tantalum Cladding from Near the Midlength of the Unirradiated Source

300X

INSIDE DIAMETER

300X

CENTER OF WALL

300X

OUTSIDE DIAMETER

Fig. 6. Photomicrographs of Typical Areas of the Tantalum Cladding from
Near the Midlength of Source SO-1915

as a result of interaction with the flowing primary-system sodium. The pitting might have occurred after the source was removed from the reactor, during storage, or during sample preparation. However, because the temperature of the source was probably less than 150°C during the short storage period, the pitting probably occurred while the source was at higher temperatures in the reactor. No evidence of platelets near the surfaces or at the grain boundaries was observed. Such platelets are characteristic of oxide, nitride, or hydride precipitates in the microstructure of tantalum. The optical metallography did show that the grain size of the cladding of the irradiated sources was much smaller than that of the unirradiated source. Because there is no apparent explanation for grain refinement as a result of residence in the reactor, it is speculated that the difference in grain size can be attributed only to fabrication differences in the original material.

C. Density Determinations

The results of the density determinations are given in Table III.

TABLE III. Density of Samples of Tantalum Cladding

Source Number	<u>Distance from the Bottom of the Source, in.</u>		
	13	7	1
Unirradiated	16.67	16.68	16.69
SO-1915	16.44	16.36	16.48

The density changes are listed for the various distances from the bottom of the source and are plotted in Fig. 7. Control samples from the unirradiated source were measured for density by using the same apparatus and during the same session as the samples from SO-1915. Although the accuracy of the density apparatus is ± 0.005 with secondary standards, the accuracy of any density measurement depends on the configuration and size

Fig. 7. Density of the Tantalum Cladding and the Percentage of Density Change as Functions of Distance from the Bottom of the Source

of the sample measured. Another factor affecting the accuracy of these density measurements is the gamma-and beta-decay heating of the sample. No significant decay heating in these samples was noted, because the temperature of the immersion bath did not increase during long sequences of making measurements on the irradiated samples. In addition, determinations were made several times in different sequences, with no changes in the results. A rough hand calculation of the beta heating of these samples indicated that each sample would produce about 0.02 W of heat 30 days after the source was removed from the reactor.

The lattice parameter of tantalum is also dependent upon the impurity content of the metal. Gebhardt³ has shown that, in unirradiated tantalum, the lattice parameter changes from 3.302 Å for high purity tantalum to 3.306 Å for tantalum containing 1 at. % (0.09 wt %) oxygen. This change in lattice parameters and increase in oxygen content reduces the theoretical density of the tantalum from 16.689 to 16.536. If the nitrogen and oxygen content of the irradiated tantalum is assumed to change the lattice to the same extent as an equal amount of oxygen would change it, the impurity content of the tantalum would account for a $\Delta V/V$ of 0.92%. The maximum density change observed was 2.04% near the midlength of the source, whereas a minimum density change of 1.20% was observed near its bottom. These data indicate that pickup of impurities from the sodium could contribute significantly to the observed swelling of the tantalum cladding in EBR-II. In contrast, a comparison of the density changes and the shape of the relative axial flux indicates that total swelling is a function of the neutron exposure the cladding received. If a $\Delta V/V$ of 0.92% is subtracted from the maximum total swelling obtained from these density measurements a maximum swelling of 1.12% can be attributed, by these density measurements, to the irradiation-induced swelling.

D. Microhardness Measurements

The results of microhardness measurements on samples of the unirradiated source, SO-1912, and SO-1915 are given in Table IV. The results listed for the samples from SO-1915 are the average values for a microhardness traverse from the outside diameter to the inside diameter of the

TABLE IV. Microhardness of Samples of Tantalum Cladding
(in kg/mm^2)

Source Number	<u>Distance from the Bottom of the Source, in.</u>		
	13	7	1
Unirradiated	98	82	78
S0-1912	---	277	---
S0-1915	217	302	311

tubes in increments of about 0.005 in., starting about 0.002 in. from the outside surfaces. The hardness measurements were very uniform across the wall thickness--within 5%. These results indicate the absence of large changes in the interstitial content in the material from the inside to the outside of the tubing.

Data on unirradiated tantalum indicate that microhardness is dependent upon its impurity content. Data by Gebhardt³ show that tantalum containing 1 at. % (0.09 wt %) oxygen has a microhardness of about 260 kg/mm^2 . Seghezzi⁴ has shown that the microhardness of tantalum containing nitrogen varies with the nitrogen content at almost the same rate as the microhardness of tantalum containing oxygen varies with the oxygen content. If the nitrogen and oxygen contents are combined and the combination is assumed to change the microhardness at the same rate as does oxygen, the microhardness of source S0-1915 due to the combined gas content would be about 260 kg/mm^2 . The sample from the same location as the sample analyzed for the gas had a microhardness of 302 kg/mm^2 . These data indicate that most, if not all, of the changes in microhardness can be attributed to impurities picked up from the EBR-II sodium rather than to irradiation damage.

E. Diameter Measurements

The postirradiation measurements of diameters on all three irradiated sources and the control source are given in Table V. None of the diameters are larger than the maximum specified diameter (0.916 in.)

TABLE V. Diameter Measurements of Sources

Source Number	Distance from the Bottom of the Source, in.				
	13	10	7	4	1
Unirradiated	0.912	0.912	0.914	0.914	0.914
SO-1911	0.914	0.916	0.916	0.915	0.913
SO-1912	0.916	0.915	0.915	0.915	0.913
SO-1915	0.915	0.914	0.913	0.914	0.915

for as-fabricated sources. However, because preirradiation measurements were not made on these sources, whether any diameter change actually occurred as a result of irradiation is still uncertain.

IV. CONCLUSION

The tantalum cladding from sources irradiated in EBR-II have shown appreciable changes in microhardness and density, and there is evidence of possible corrosion at the outer surface of the cladding. Unfortunately, the tantalum cladding was not well characterized before its use in the reactor; therefore, the conclusions drawn from the results of these examinations are only qualitative. An unirradiated source fabricated at the same time the irradiated sources were built was used as a control and examined, but metallographic examinations revealed that it may not be typical of the irradiated sources.

Another factor that complicates correlating the data from the examination of the tantalum cladding is that the sources are not irradiated under conditions typical of the EBR-II environment. The sources spent most their time in the reactor in the photoneutron-source assembly, in which they were surrounded by a beryllium cylinder. The beryllium acts as a moderator; thus the tantalum is exposed to a neutron spectrum softer than the nominal EBR-II flux spectrum. This factor confuses still further the uncertainty of the fluence values reported for the sources.

The results of the examination indicate that:

1. The tantalum cladding has not been severely attacked by the EBR-II primary-system sodium, although some pitting was observed.

2. Significant swelling occurred as a result of irradiation of the cladding. The density changes appeared to be a function of the fluence, because the maximum change was observed at the midlength and lesser changes occurred at both the top and bottom end. The amount of swelling (reduction in density) that can be attributed to irradiation damage is unclear because part of the change in density appears to have resulted from chemical changes that occurred while the source was in the reactor.

3. The mechanical properties of the tantalum cladding were altered, as evidenced by the increases in microhardness. The changes appear to have resulted from a combination of irradiation effects and chemical changes brought about by the takeup of impurities. Increases in the oxygen and nitrogen content of the tantalum cladding may have caused much of the observed change in microhardness.

4. Irradiated tantalum can pose significant problems in postirradiation handling because of its high-energy gamma activity. These problems can become very significant if small particles are generated during sample preparation.

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