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Metallurgical Tests on Zirconium-Clad Zr-U Alloy Plates Submitted to Hanford for Irradiation

Program 6.1.2a

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AEC RESEARCH AND DEVELOPMENT REPORT

A. Summary of Tests

Four plates containing enriched material were submitted for examination, two each of dimensions approximately .15" x .85" x 7.33" and .15" x .85" x 2.02". Specimens were numbered #5 and #7, and #9 and #10. Cover plates and picture frames were rolled from ingots of Foote crystal bar melted by arc under a coated electrode, except for #7 which had cover plates made of M.I.T. extruded crystal bar. Full details of fabrication process are given in reference (1).

In addition, a sandwich plate containing natural uranium-zirconium alloy clad with Bureau of Mines zirconium was submitted for corrosion tests. This specimen is listed as one of the short pieces of Item #3 in reference (2). In all specimens the "meat" contained Bureau of Mines sponge metal.

Enriched specimens were polished metallographically along the edges, and the following tests were performed:

I. Metallographic examination. All bonds along edges of enriched specimens were examined at a magnification of 90-X before and after water autoclave corrosion test described below. Possible small defects were marked with a scratch for identification.

II. Corrosion Tests. (Paragraphs 5 to 7 of reference 3). All specimens were placed in aluminum holders and autoclaved in distilled water at 150-160°C for 63 hours, weights noted before and after test.

The specimen containing natural uranium was then cut to expose the "meat" and one section was boiled in aerated distilled water for 32 hours, while another was tested in contact with aluminum for 48 hours at 157-167°C in a water autoclave.

References: (1) L. W. Eastwood (B.M.I.) to E. G. Ashcraft (A.N.L.), dated 3/21/49.
(2) Ibid., dated 3/24/49.
(3) ANL-HE-47, S. Naymark to distribution, dated 4/7/49.

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III. Bond Cleavage Test. Portions of the cut specimen were placed in a vise and the bond was cleaved by using a sharp chisel.

IV. Physical Property Tests on Enriched Specimens. Complete dimensional and profile measurements were made. Rockwell 15-T superficial hardness survey was made. Density of specimens by loss of weight in carbon tetrachloride was determined. Data on these physical tests will be the subject of a separate report.

B. Results

I. Metallographic Examination of Bonds Before Autoclave Test

Due to the awkward size and shape of the specimens, metallographic preparation was done by hand, using 250, 400 and 600 grit papers, followed by hand lapping with relevigated alumina.

In general, the bonds were all made visible by an intermittent line of inclusions along a wavy interface, (see Figures 1, 2 and 3). The zirconium cladding material was not completely sound, but small voids and rolling defects were observed in the surface of the specimens, the largest of which is recorded in Figure 3. All visible defects in bonds and in cover material were marked with a needle scratch for reference after autoclave tests. /end

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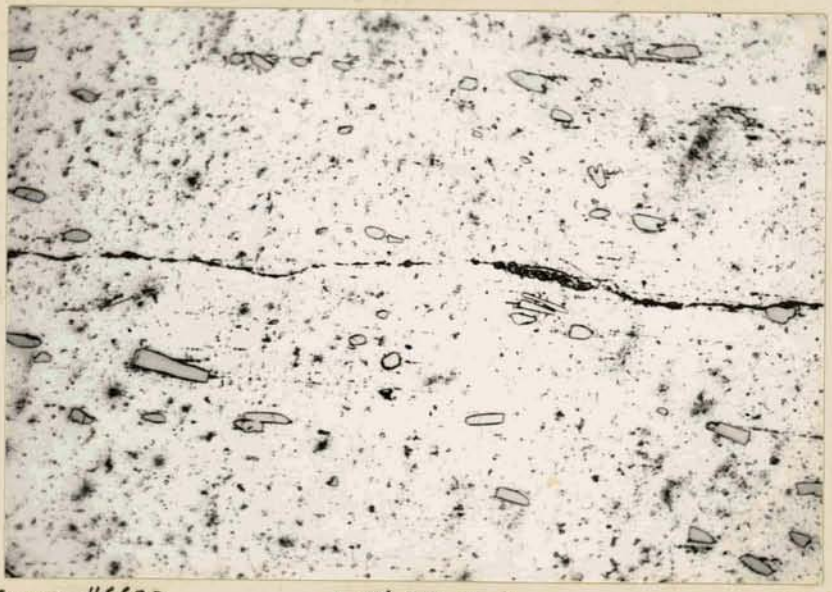
Figure 1. Specimen No. 2124-1

Macro #6631 0.5% HF etch 100-X

View of Bureau of Mines clad specimen bond, showing extensive inclusions supposed to be carbides. Structure is very similar to arc-melted crystal bar material used for the Hanford and Oak Ridge test specimens. (Compare with Figure 3.)

Figure 2. Specimen No. 2124-1



Macro #6630 0.5% HF etch 500-X

Higher magnification of area shown in Figure 1. Note rather vigorous attack of HF on bond inclusions, as compared with negligible attack on matrix carbides.

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Figure 3. Specimen No. 2124-10

Macro #6591 As Polished 100-X

View of polished edge on Hanford test specimen #10, fabricated from arc-melted Foote crystal bar. Polishing is not optimum, due to awkward shape and size of piece, but matrix inclusions (carbides) are visible as well as a discontinuous line of inclusions at bond. Note macro voids visible in the edge of cover material above (diagonal mark at upper right is a needle scratch). These are rolling defects, the only serious ones observed on any of the Hanford specimens. Recent tests indicate that they contain a black material which is not yet identified.

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II. Autoclave Test on Enriched Specimens

In this test the samples were placed in aluminum holders in a stainless steel autoclave, which was then filled approximately 8/10 full with distilled water. A sample of the water was taken and stored in a covered beaker until the end of the test. To the autoclave was connected a copper tube, leading to a pressure gauge. The system was placed in the furnace in such a way that the gauge was outside.

The thermocouple recorded temperature of the furnace during the test range from about 150-160°C. The thermocouple, however, was near the top of the horizontal furnace and the autoclave was placed at the bottom. Pressure readings increased for the first 19 hours to a value of 50 psi and remained in the range of 48-51 psi until the duration of the test, 63 hours. Results of test are tabulated below:

Weights in Grams

	Before	After	Remarks
Samples			
5	101.483 gms	101.483 gms	Very little oxide
7	102.416	102.417	Aluminum oxide
9	28.107	28.108	Clean
10	27.964	27.964	Clean
Natural			
large	46.509	46.509	Clean
small (smooth)	8.092	8.092	Clean
small (rough)	7.934	7.934	Clean
Holders			
5*	336.250	336.500	Considerable red stain
7*	336.175	336.485	Slight red stain
9	163.063	163.116	Red stain
10	163.171	163.242	
no number	179.460	179.498	

* ±0.005 gm: All other wts. to 0.0005 gm.

The stainless steel autoclave was contaminated at the beginning of the test with a white material presumed to be alumina. This doubtless accounts for general superficial staining of the specimens shown in Figures 4 and 5. In addition, all samples showed a thin tannish film probably indicating the presence of a slight oxide film. The aluminum holders showed considerable pitting attack, particularly adjacent to areas of contact with the stainless steel and in some

cases adjacent to the samples (Figure 6). A reddish stain was apparent on some of the sample holders. This material has not been identified. It yielded quick qualitative tests for iron and copper.

The water conductivities before and after tests were 69,800 and 22,500 ohms/cm^3. The pH before and after test was 5.1 and 5.9 respectively. It should be noted that the water sample taken before test had been allowed to stand in somewhat of a restricted contact with the air for 65 hours before its pH and conductivity were measured.

No pitting was observed on any of the sandwich specimens. The bond lines proved to be stained by the test (Figures 4, 7). Some boundary attack around bond inclusions seems to be responsible for this condition; however, penetration was not great (Figures 7, 8 and 9).

Figure 4. Specimen No. 2124-10

Micro #6592 As water autoclaved 100-X

Micro of area shown in Figure 3 after water autoclave test in contact with aluminum. General staining is alumina. Water seepage from macro defects is noted. Staining along bond is further analyzed in Figures 7 to 9.

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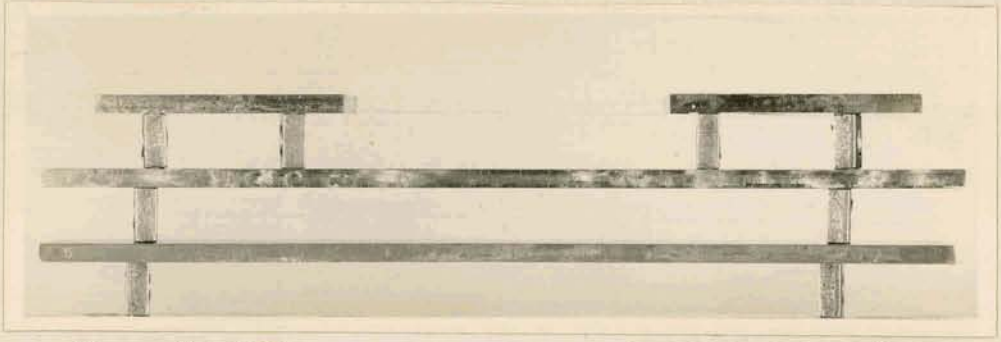
Figure 5.



Macro #M-4437 Approximately 2/3-X

Macro appearance of specimen edges after water autoclave test in aluminum holders. Small vertical scratches were made with scratch-all before test to identify inclusions or voids in bond. No reaction was noted at these points.

Figure 6.

Macro #M-4436 Approximately 2/3-X

Macro appearance of outsides of 2-S aluminum holders. Holders were badly pitted by the autoclave test, and produced considerable alumina.

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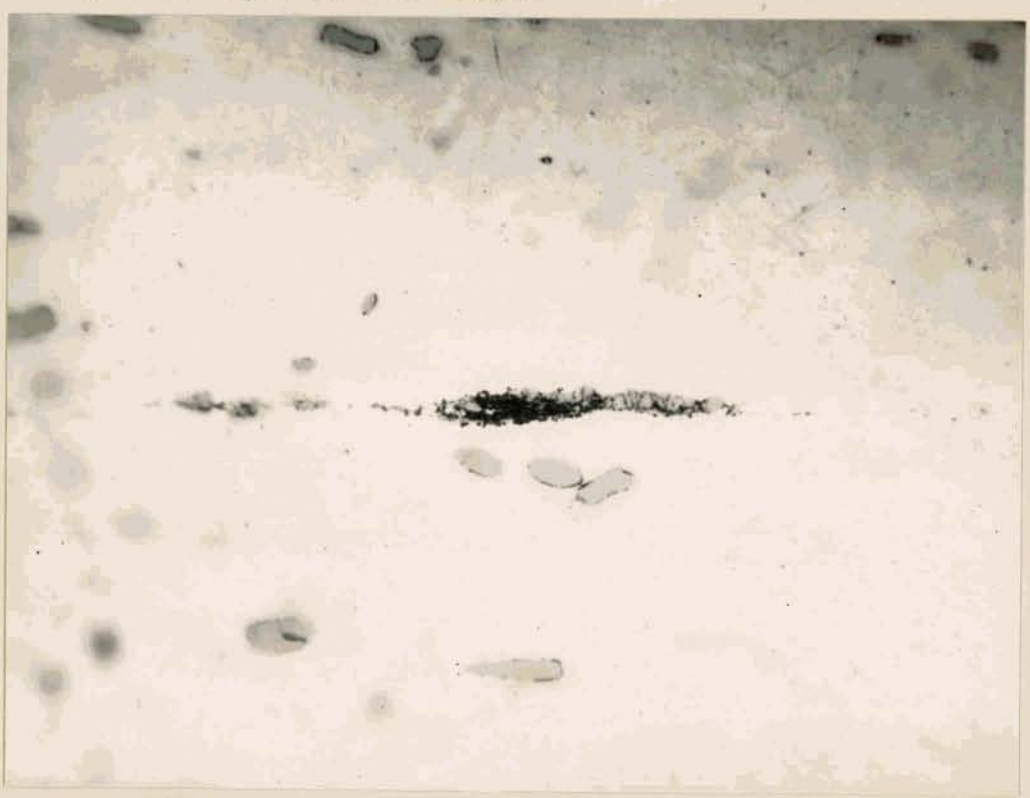
Figure 7. Specimen No. 2124-10

Bond

Micro #6593 1500-X

Bond staining after autoclave test, shown at high magnification. This area is characteristic of the maximum effect observed.

Figure 8. Specimen No. 2124-10



Bond

Micro #6622 1500-X

Same area as Figure 7 after 3 minutes of light polishing with Linde B (.1 micron) powder on AB Microcloth. Staining has disappeared but boundary corrosion at all inclusions is still visible. Loosened inclusions at bond have been pulled out.

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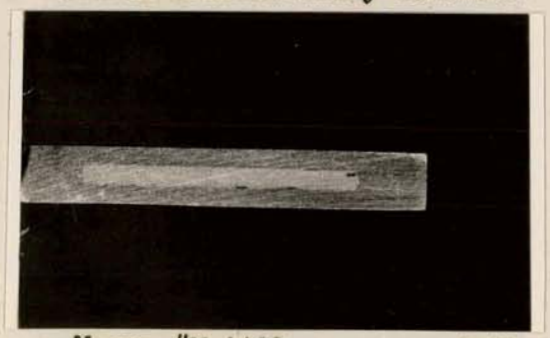
Figure 9. Specimen No. 2124-10

Bond

Micro #6621 As polished 1500-X

Same area as Figures 7 and 8 after additional polishing totalling 10 minutes. Boundary corrosion at inclusions has disappeared in areas away from bond. It is beginning to disappear around bond inclusions, but a void appears where particles have been pulled out. Total depth of metal removed is less than a micron. Total length of main defect is approximately 25 microns.

Figure 10. Cross Section,
Natural Uranium Alloy Sandwich



Macro #M-4439 2-X

Differential surface oxidation between meat and cladding of cut sandwich specimen after water autoclave test. No swelling occurred. Four macro defects in meat are visible.

III. Corrosion Tests on Sectioned Specimen with U^{238} Meat

Autoclave and aerated boiling water tests on sections with exposed meat (natural uranium alloy) produced no pitting or swelling. The only observable effect was a slow surface oxidation which produced a bronze diffraction layer on the zirconium and a gray layer on the meat. Neither of these layers showed any tendency to flake (see Figure 10).

The autoclave test was run in an aluminum (2S) can placed in a stainless steel autoclave. The can was washed, it and the sample were degreased in carbon tetrachloride, then acetone, blotted on Kleenex, dried and weighed. Distilled water was put in the can to cover half the sample. The bomb contained distilled water to within about a quarter inch of the top of the can. The water in the bomb was heated to boiling, and the cover put on while still boiling.

After a forty-eight hour test at 162°C (furnace range 157-167), no pitting was evident, although a small amount of a white material, apparently aluminum oxide, showed on one end of the specimen. The can was covered with a white coat, probably aluminum oxide; no pitting was evident.

Weights in grams

	Before	After	Gain
Sample	10.3971	10.3975	0.0004
Can	13.7209	13.7490	.0281
Water resistivity	203,000 ohms/cm^3	35,400 ohms/cm^3	

The water conductivity was measured in the steel bomb. After test, the can was two-thirds full of water (covering the sample completely). This was poured into the bomb with the rest of the water and the conductivity of the composite measured.

IV. Cleavage Tests

Sections of an autoclave-tested specimen having U^{238} in the meat were submitted to a chisel test. This is a simple but quite rigorous method of testing a metallurgical bond. It turned out to be rather difficult to start a split along the bond, half a dozen heavy blows being required to do so. Once started, however, the cleavage progressed fairly readily in three out of four tests. The last test produced tearing of the sandwich material. Bonded surfaces had a bright pebbled appearance indicating complete contact of cover metal, with the exception of one spot which had a brown discoloration running from edge to corner of meat. This apparently is a perforation which was entered by autoclave water (Figure 11). Edges of this specimen material had not been prepared metallographically for careful examination before autoclave test, and it is felt

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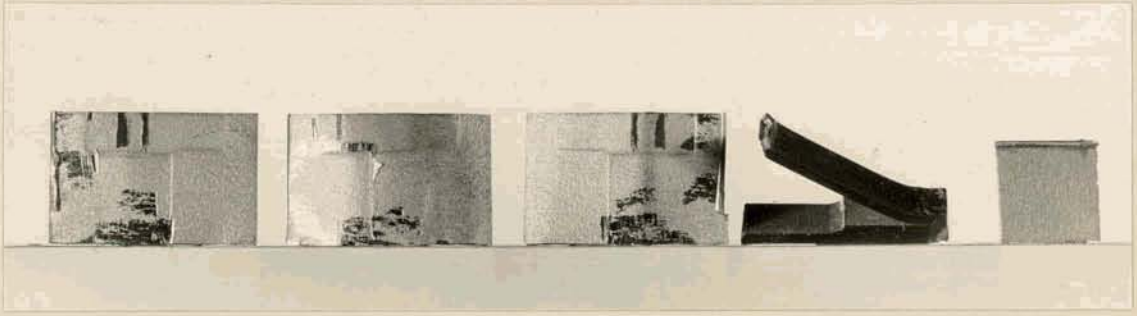
that this defect would certainly have been noted if it had been so examined. We are also informed by members of the B.M.I. staff that this specimen could be classed as definitely worse than the enriched specimens from the fabrication technique viewpoint, as it represents quite an early attempt to bond sandwich type assemblies.

Figure 11. Cleavage Tests on Natural Uranium Alloy Sandwiches



Macro #M-4435 Approximately 1-X

Macro of two chisel test specimens after bond cleavage test. All split bonds were bright and clean except for macro defects in meat (1), and brown stain (2) which resulted from autoclave water penetration at a faulty bond on one specimen. The other specimen cleaved with difficulty and finally failed by tearing. Specimens were from a natural uranium sandwich fabricated in initial process experiments.

C. Conclusions

Results of the corrosion tests performed on enriched specimens and on specimens with exposed meat lead us to conclude that they should readily withstand the corrosive conditions imposed upon them by the proposed Hanford pile experiment.

From the metallurgical standpoint, however, we feel that cleaner zirconium and a cleaner bond between cover and picture frame must be sought before the assembly will become satisfactory for performance under conditions involving high temperature, high pressure water.

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