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ZRCLAD ZIRCONIUM-URANIUM ALLOY ROD FOR HEAT THRU-PUT TEST

by

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INTRODUCTION

The purpose of this work was (1) to investigate the possibility of producing zirconium-clad zirconium-uranium alloy objects by casting the core alloy directly into zirconium and zirconium-3 w/o tin jackets, producing a diffusion bond at the jacket-core interfaces, and (2) to produce small scale heat thru-put test specimens by this method.

The test specimens used in this work are shown in Figure 1. These specimens were to be used for heat transfer studies between core and cladding. Their requirements were as follows:

a. Core and cladding dimensions should be accurate.

b. The core should be without internal voids.

c. The bond between the core and cladding should be as nearly perfect as possible.

Method

The following tentative sequence was selected as an approach to the solution of the problem:

a. Vacuum melt and cast zirconium-4 w/o uranium alloy into a heated zirconium (or zirconium-3.0 w/o tin alloy) jacket mold with one end cap shrink-fitted in place to form the mold bottom.

b. Diffuse core to mold and cap.

c. Radiograph.

d. Remove casting top, containing shrink hole, and machine to fit upper cap.

e. Shrink or press fit upper cap to cast assembly.

f. Radiograph.

g. Diffuse upper cap to assembly.

h. Machine to final dimensions.

Equipment Used

Melting and Diffusing Furnace

The furnace used for melting the zirconium-uranium alloy, for heating the mold and for diffusion of the specimen ends and jacket is shown in Figure 2. The zirconium mold assembly is shown in Figure 3. The furnace assembly was mounted on a vacuum system capable of maintaining an indicated vacuum of between 10^{-5} to 10^{-4} mm Hg throughout the process. The vacuum gauges were mounted on the vacuum chamber. Much higher pressures would have been found in the mold heating zone and at the upper end of the furnace tube had it been possible to make measurements there.

The melting zone of the furnace was heated by induction to a 0.025" thick tantalum heater around the crucible. Two split cylinders of 0.005" molybdenum were placed concentrically around the heater. These reflected the heat back toward the heating zone and kept the Vycor tube well below its softening temperature, which was about 850°C. This furnace was found to be an efficient heating arrangement. Nine kilowatts motor-generator output heated the crucible and charge to 1850°C.

Slip-cast thoria crucibles were used. These were approximately 2-1/4" in diameter at the top, 1-7/8" in diameter at the bottom and 3" deep. Each had a conical bottom for clean drainage. Heavy walled crucibles, about 1/4" thick, were used at first. These were extremely susceptible to stress cracking by nonuniform heating and cooling. The wall thickness was reduced to approximately 1/16". The thin walled crucibles, although fragile to handle were found to allow high heating and cooling rates and were strong enough to hold a full charge of metal.

The tapered bottoms required that a special pedestal be used to support the crucible. These were machined from zirconia brick and were lined with a slip-cast thoria funnel. The lower end of the funnel was fitted into the mold and formed a hot top for the casting when filled with metal.

The furnace charge was bottom poured by lifting a thoria stopper rod which plugged a lapped hole in the crucible bottom.

The mold heater consisted of a 1-9/16" inside diameter alumina tube wound with 25 feet of 0.030" platinum wire. This was placed inside a fused silica tube which supported the melting zone assembly. Magnesia spacer plates were machined to interlock with the various parts of the upper and lower assembly, holding them rigidly in place. Magnesia plates were also used to insulate the mold heater from the water cooled pedestal in the vacuum chamber. Power was supplied to the mold heater by a 20 amp 220 volt variable transformer through copper rods extruding through rubber glands in the lower vacuum chamber cover. The heater would stand about

17 amperes continuously; however, when the castings were poured it was necessary to reduce the current to prevent burn-out of the element.

Several difficulties were experienced in using the equipment. The entire assembly was found to expand about 3/8" when heated. A tantalum wire linkage was used between the stopper rod and manipulator rod. Clearance and a sliding joint were provided in the upper MgO positioning plate to allow for the difference in expansion of the alumina mold heater tube and the quartz supporting tube.

A reflecting shield was mounted on the stopper rod manipulator to keep the upper silicone rubber gasket from being charred by the heat of the melting zone.

It was necessary to separate the zirconium mold from the magnesia and zirconia ceramics. Tantalum was found to be satisfactory. The upper end of the mold which penetrated into the melting zone was heated to the fusion point. This effectively "hot topped" the casting and helped prevent formation of a shrinkage pipe.

Experimental Work

Table I shows melting and diffusion welding data for 11 heats. Six of these were poured into crystal bar zirconium molds. Three were poured into 3% tin-zirconium alloy and two were poured into water cooled copper molds.

Interrelated factors which were found to affect the casting quality and the bond were:

1. Outgassing the charge and mold before melting.
2. Pouring temperature.
3. Size of crucible tap hole.
4. Mold temperature at time of pour.
5. Condition of inside surface of mold.
6. Time and temperature of diffusing core and mold tubes.
7. Time and temperature of diffusing end caps.
8. Degree of negative clearance allowed for the end caps.

Outgassing

Some difficulty was experienced in maintaining a vacuum of between 10^{-5} and 10^{-4} mm of Hg at the high pouring temperature. In most of these heats the furnace assembly was set up the day before melting and was

Table 1

MELTING AND WELDING DATA, HEAT THRU-PUT TEST SPECIMENS

Melt No.	Zirconium (Grams)	Uranium (Grams)	w/o U	Pouring Temp. (°C)	Pouring Vacuum (mm Hg)	Outgassing Remarks	Mold Material	Mold Temp. (°C)	Diffusion: Mold Tube	Diffusion: Caps	Remarks
1	119.916	5.468	4.36	1860	5.2×10^{-5}	Not outgassed before melt. Start to finish 2 Hrs.	Crystal Bar Zirconium	980	2 Hrs. 900°C	2 Hrs. 900°C	No pipe. About 90% bonded. Sectioned.
2	159.098	7.065	4.25	1850	6.5×10^{-5}	Not outgassed before melt. Start to finish 2 Hrs., 45 min.	Crystal Bar Zirconium	1226	2 Hrs. 1200°C	2 Hrs. 1200°C	Stopper rod was not sealed. Shot dribbled into lower end of mold. Bond poor.
3	141.445	7.175	4.83	1800	1.2×10^{-5}	Outgassed. Charge and Mold, 2 Hrs. at 850°C	Crystal Bar Zirconium	900	2 Hrs. 850° - 900°C	2 Hrs. 850° - 900°C	Casting good. Bond poor, little diffusion. Some laps.
4	152.669	7.649	4.74	1830	1.2×10^{-4}	Outgassed 2 Hrs. at 800°C	Crystal Bar Zirconium	1150	Mold Heater Burned out	2 Hrs. 950° - 1000°C	Casting showed some lapping. Diffusion good except at laps.
5	153.037	7.215	4.50	1850	4.5×10^{-4}	Outgassed 1 Hr. at 1000°C	Crystal Bar Zirconium	1200			Stopper rod broke.
12	157.693	7.112	4.31	1860	8.0×10^{-5}	Outgassed 1 Hr. at 1000°C	Crystal Bar Zirconium	1155	5 min. 1155°C Heater burned out	2 Hrs. at 1200°C	Casting sound. Well bonded.
6	155.303	7.916	5.30	1850	1.8×10^{-4}	Outgassed 1 Hr. at 975°C	Zr-3 w/o Sn	1000	2 Hrs. 1000°C	Upper Cap Hot	See Note(1)
7	152.701	7.697	4.80	1820	6.2×10^{-5}	Not outgassed before melt.	Zr-3 w/o Sn	1000	2 Hrs. 1000°C	Upper Cap Hot	See Note(1) Metal blew back into crucible.
10	154.165	7.841	4.84	1840	1.4×10^{-4}	Outgassed 1 Hr. at 1000°C	Zr-3 w/o Sn	1000	2 Hrs. 1000°C		Footnote(1)
8	646.713	29.276	4.21	1900	3.4×10^{-4}		1-1/8" Dia. Cu W.C.	---	---	---	See note(2) Casting was not sound.
11	666.788	29.209	4.21	1860	2.0×10^{-4}	Outgassed 3 Hrs. 975° - 1000°C	Copper Water Cooled	---	---	---	Slight secondary pipe at lower end of casting.

(1) An evolution of gas occurred in each case when molten zirconium at 1850°C was poured into Zr-Sn alloy molds.

(2) Melts 8 and 11 were poured into a water cooled copper mold.

outgassed from one to four hours at approximately 1000°C. The power to the heaters was then turned off and the furnace pumped over night.

Pouring Temperature

Laps and poor bonding were experienced when the pouring temperature was 1800°C. Above 1850°C erosion of the crucible by the molten metal became severe. This was particularly noted in melt number 8 in which the temperature was accidently raised to 1900°C. The casting was not sound and the quality of the metal was poor.

Pouring Rate

Pouring rate is controlled by the size of the crucible tap hole. The first stopper rods available were 1/4" to 5/16" in diameter, requiring that a 3/16" hole be used in the crucible bottom. This was too small and caused laps in the first four castings made. One-half inch diameter stopper rods became available for the later castings allowing 3/8" diameter holes to be used in the crucible bottom. The results were more satisfactory.

Mold Temperature

The temperature of the mold heating zone was read by means of a platinum-10% rhodium thermocouple inserted through the wall of the mold heater. Best results were obtained when temperatures read on this thermocouple were above 1100°C. When the 1800°C alloy was poured into the mold at 100°C, it was necessary to interrupt the power to the mold heater momentarily to prevent heater burn-out.

Condition of Inside Surface of the Mold

A uniformly roughened surface was found to give better wetting and bonding than a smooth surface. Figures 3 and 4 show the type of surface used on the inside of the mold and lower cap. This was produced by a 1/32" radius boring tool. For melt #12 the lathe was set at 80 threads per inch; this produced good results. Thirty threads per inch, used on melt #1, produced too rough a surface and there was incomplete penetration into the valleys between the threads.

Diffusion of Cores, Tubes and Caps

The ends of the specimens were fabricated as caps which were pressed onto the ends of the mold tube. The lower cap formed the bottom of the mold. The upper cap was placed on the mold after filling it with the core alloy. These caps were machined to a negative fit of 0.0002" to 0.0004". The caps were heated in an oven to about 250°C and the tubes were forced into the caps using a hand arbor press. Hammering on the upper end of the ram assisted in seating the tubes.

After assembly the specimens were set into the furnace with the cap in the heating zone. They were then heated to the temperatures indicated in Table I. Diffusion across the interface occurred at about 1100°C. The weld was improved at 1200°C and 1300°C. An objection to this method of welding is the growth of very large grains. Figure 5 shows the specimen produced by melt #12 after diffusion of the ends.

Finish Machining

After assembly and diffusion, the specimens were straightened using an arbor press, balancing rolls and indicator. They were then machined to final dimension on centers on a lathe. Figure 6 shows the finished machined specimen.

Inspection

X-ray inspection was made of all castings. The first 10 castings were sectioned longitudinally and metallographic examination was made of the bond produced. Sectioning of finish specimens was not possible and dependence was placed on the radiographs. Figure 7 shows a well diffused bond between the core alloy and jacket, while Figure 8 shows a specimen in which the core alloy did not penetrate into the valleys between the threads.

Specimen #12 was finished and submitted to the Reactor Engineering Division for further evaluation.

Discussion of Results

A. Pure zirconium clad specimens: The method was found to be feasible when a crystal bar zirconium mold was used as the cladding material. If quantities of zirconium clad specimens were required considerable modification of the technique would be necessary. One possibility might be casting core alloys into zirconium molds and bonding by further fabrication processes such as rolling or extrusion. An advantage of this would be the elimination of open fabrication and machining in working and cladding enriched alloys or alloys with high alpha activity.

B. Zirconium-3 w/o tin clad specimens: The technique was not found to be satisfactory for casting into zirconium-3w/o tin alloy jackets. Bonding of the core and cladding was not satisfactory with the tin-zirconium alloy. The castings all showed pockets which had the appearance of "blow holes." Satisfactory melting of zirconium-uranium alloy in the equipment used was dependent upon maintaining a good vacuum. The vapor pressure of tin is given as follows:

Table II

VAPOR PRESSURE OF TIN[1]

Pressure in mm	1	10	40	100	400	760
Temperature °C	1492	1703	1855	1968	2169	2270

[1] Vapor Pressure of Inorganic Compounds, Handbook of Chemistry and Physics - 33rd Edition (1951-1952) p. 1952.

The high vapor pressure resulted in a boiling action when the melted alloy was poured into the 3% tin-zirconium alloy molds.

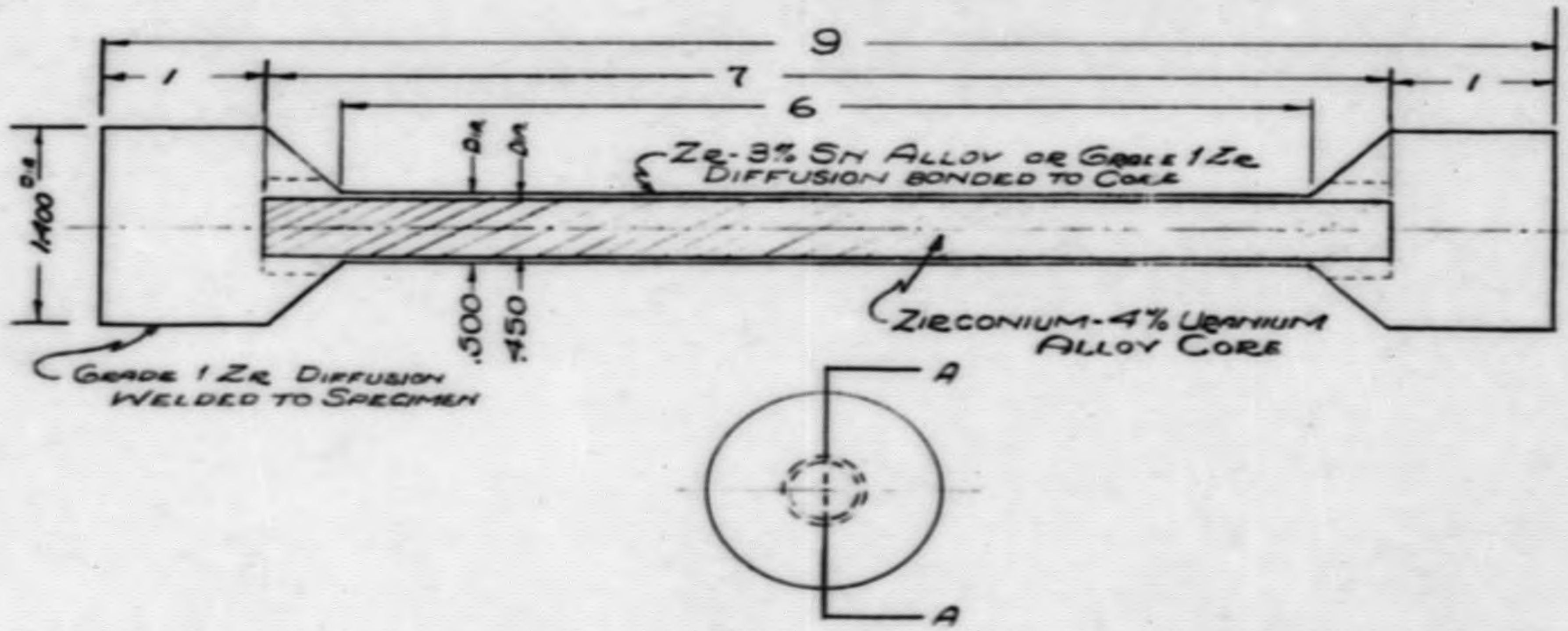

FIGURE 1: SMALL SCALE LOOP TEST SPECIMEN

ABS
4-3-52

MY 1417A

FIGURE 2
SCHEMATIC FURNACE ASSEMBLY
DRAWING

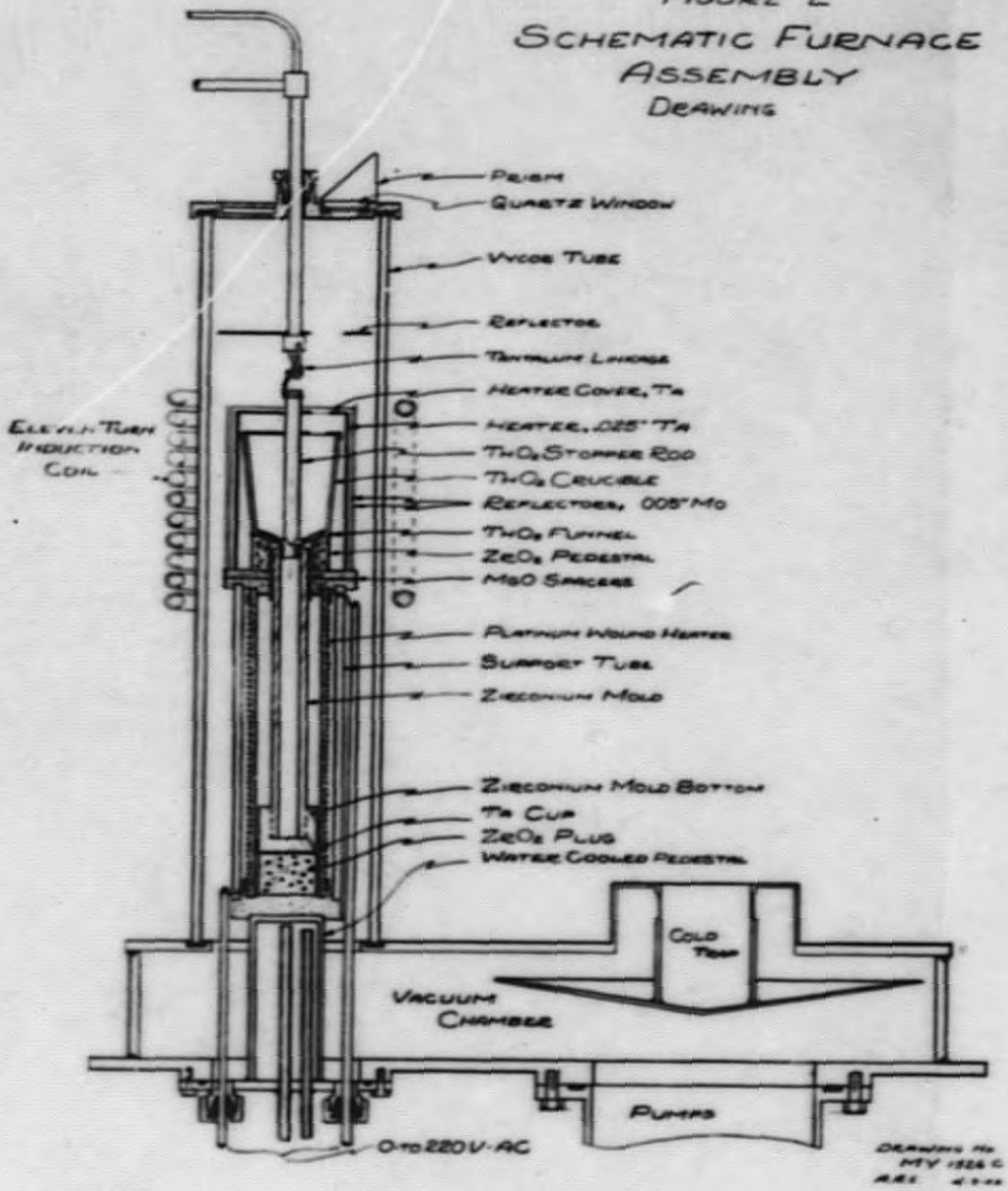

Figure 3. Zirconium Mold before Fitting Lower Cap

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Figure 4. Lower Cap Showing Serrated Surface

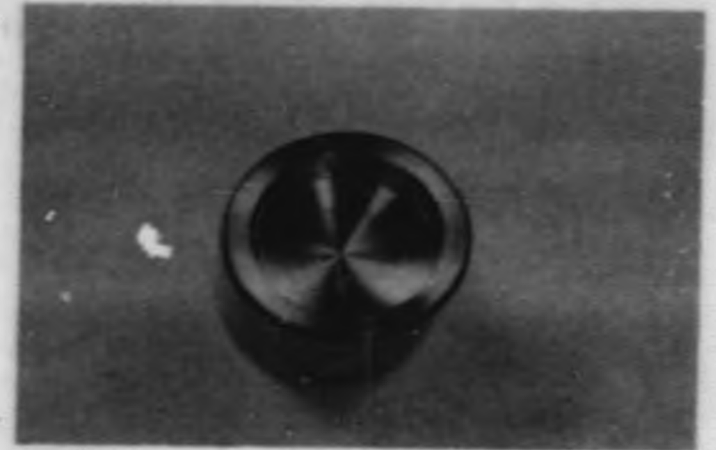

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Figure 5. Specimen Assembly After Diffusion of End Caps

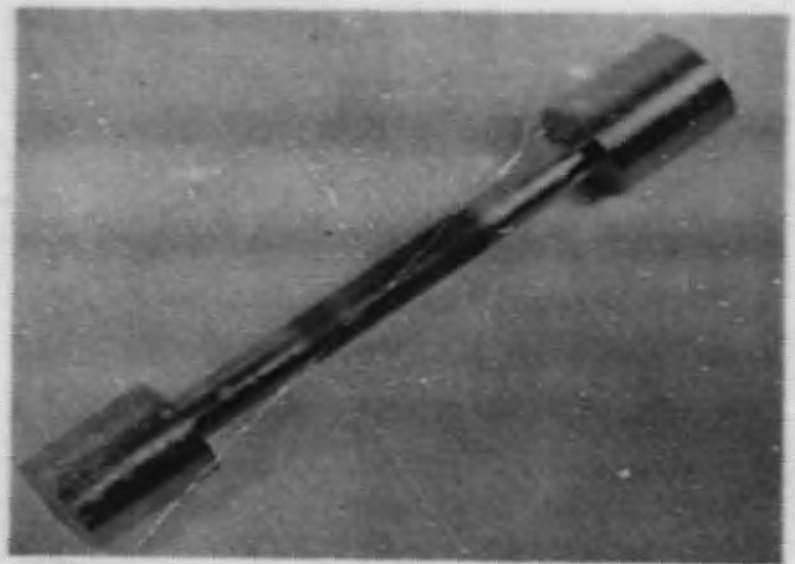

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Figure 6. Zirconium Clad, Zirconium-4.3w/o Uranium Alloy Loop Test Specimen

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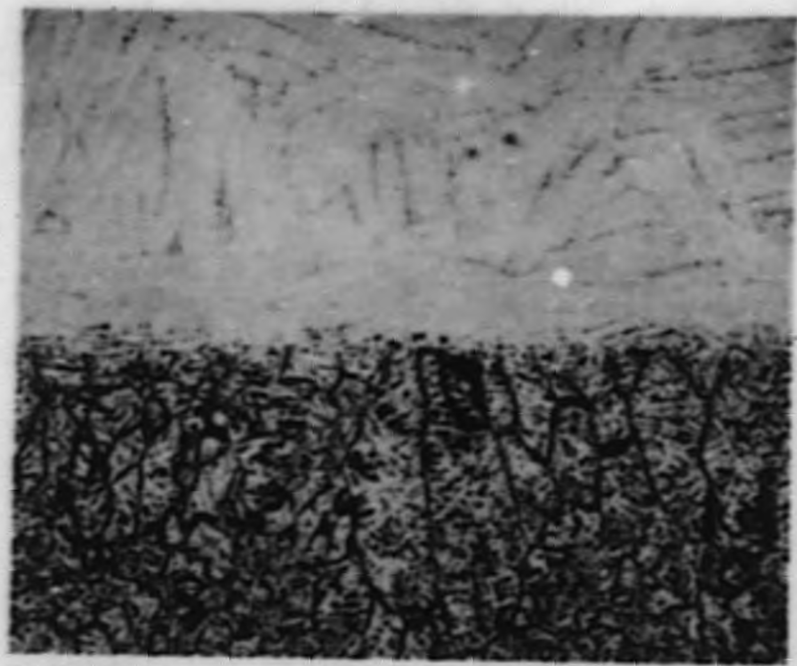
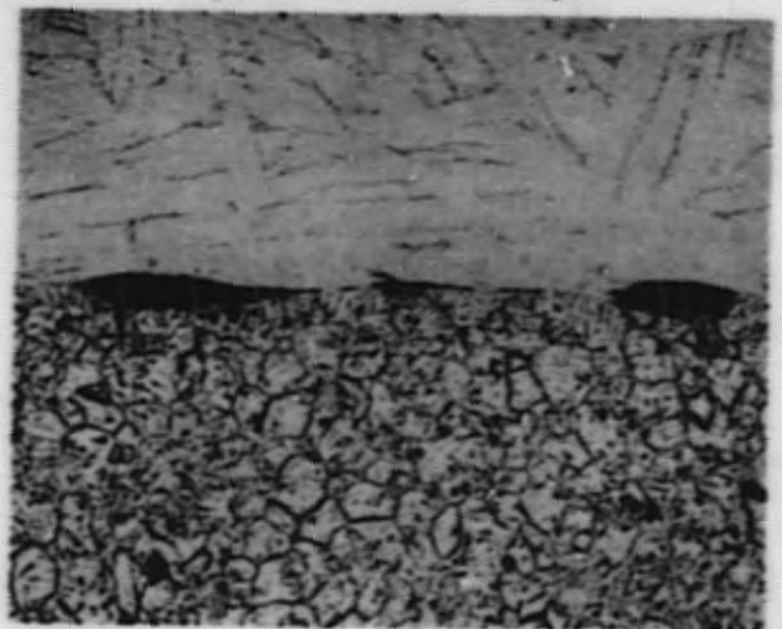
Figure 7. Diffusion Bond between Zr Cladding and Zr-U Core Alloy



M-12654 50X

Etchant - 3 grams of Silver Nitrate, 5cc HF, 92cc H_2O.

Figure 8. Imperfect Diffusion Bond between Zr Cladding and Zr-U Core Alloy



M-12653 50X

Etchant.- 3 grams of Silver Nitrate, 5cc Hf, 92cc H_2O

END