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POSTIRRADIATION EVALUATION OF ZIRCALOY-2 PRTR PRESSURE TUBES

Part I

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By

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INTRODUCTION

The Plutonium Recycle Test Reactor (PRTR) is a heavy water cooled and moderated, vertical pressure tube reactor. The primary coolant flows upward through 85 Zircaloy-2 pressure tubes at an average of 107 gpm. The nominal inlet conditions are 478 F and 1090 psig and the nominal outlet conditions are 530 F and about 1080 psig. The connections between the Zircaloy pressure tubes and the stainless steel out-of-reactor piping are bolted flange type connections with spiral-wound, asbestos-filled gaskets. This reactor represents one of the first major elevated temperature applications of Zircaloy-2 as a pressure-containing structural material. Since prior experience with Zircaloy-2 in comparable operating environments consists only of a few years experience with single tube test loops, a pressure tube surveillance program has been undertaken in conjunction with the PRTR operation. This surveillance program consists of in-reactor monitoring,[1] preirradiation testing[2] and the periodic removal of selected tubes for postirradiation destructive examination. To date, three tubes have been removed after 2, 6, and 10 weeks of actual reactor operation. This report presents the observations made on these tubes and an analysis of the effect of exposure to the reactor environment on the properties of Zircaloy-2 pressure tubes.

SUMMARY AND CONCLUSIONS

Three tubes have been removed from the PRTR for detailed post-irradiation tests and inspections. The room temperature burst properties, depth of fretting corrosion, changes in grain structure and the extent of crevice corrosion were determined.

The observations may be summarized as follows:

1. Irradiation to 10^{20} nvt (E > 1 Mev) produced a small increase in room temperature burst strength and a decrease in ductility. The uniform deformation of the specimen prior to the local bulging that

precedes failure decreased to a negligible amount at the highest exposure. One tube failed when a short crack propagated through the wall at a stress equivalent to about 85% of ultimate burst stress. The crack did not then propagate the length of the specimen under these severe circumstances which indicates that the material is still behaving in a ductile manner at room temperature.

2. Close-up visual examination revealed that the fretting corrosion marks appeared much the same as in photos taken through the borescope. Depths of penetration from standard fuel element end brackets were found up to 6 mils deep. Outer wire wraps and, occasionally, individual rod wire wraps caused similar penetrations of the tube wall. Bands on Pu-Al elements did not cause any measurable penetration of the tube wall. Increasing the width of contact area of the fuel end bracket apparently decreased the penetration.

3. The grain structure under the fretting corrosion marks was unchanged in many instances; however, local hydriding has been found in a thin layer under three of the twelve corrosion defects that were examined. Visual appearance of the corrosion mark gave no indication of the presence of hydride.

4. Crevice corrosion of the upper flange penetrated up to 4 mils. Heavy white corrosion product is present but little if any local hydriding has occurred since the first operating period.

From the above observations it is concluded that even though the continued penetration of the tube wall by fretting corrosion would be detrimental to the integrity of the pressure tubes, the general behavior of the tubes to date has been superior to that anticipated when the tubes were designed.

RESULTS AND DISCUSSION

Effect of Irradiation on the Room Temperature Burst Properties

The design requirements of the PRTR pressure tube required a reduced diameter with a heavier wall thickness at the lower end.(Figure 1).

Fabrication processes were developed whereby the major tube diameter was produced by a 45% reduction in area by Rockrite tube reducing. The tube was then annealed and the lower end was tube reduced forming the taper and the small diameter heavy wall section. To avoid excessive warpage, only one half of the tube was annealed prior to the tapering operation. The finished tube has the unusual combination of being 45% cold worked for half its length and annealed for the other half (Figure 2).

The destructive examination of the pressure tubes is part of a surveillance program to determine the effect of the total reactor environment on the tube properties. The first visual examination of the pressure tubes after the initial two-week operating period discovered fretting corrosion in all the tubes.[1] This added a second variable for the burst tests, and the program was accelerated to determine if this fretting corrosion had proceeded to a point where special precautions were required for safe operations of the reactor.

The test sections are 20 inches long overall with an unrestrained length of 16 inches between the end closures. Figure 3 shows a test assembly with end closures and thermal couples attached. The test specimen assembly and bursting is conducted under 5 feet of water. Pressure is supplied by a 30,000 psig capacity air operated, positive displacement pump.

The tube identification numbers, weeks of operation prior to removal, and the type of fuel element contained in the tube during operation are listed in Table I.

TABLE I

OPERATING DATA ON PRESSURE TUBES REMOVED FROM PRTR

Tube No.	Channel No.	Weeks of Actual Operation	Type of Fuel	Maximum Outlet Temperature
6100	1154	2	UO_2	450 F
6061	1756	6	Pu-Al	530 F
5529	1159	10	UO_2	530 F

Two sections were burst from Tubes 6100 and 5529 and a single test was performed on Tube 6061. The level of irradiation, ultimate burst strength calculated from the burst pressures, the uniform elongation and the maximum elongation are listed in Table II.

TABLE II

EFFECT OF IRRADIATION
ON ROOM TEMPERATURE BURST STRENGTH
AND ELONGATION OF ZIRCALOY-2 PRTR PRESSURE TUBES

Calculated Exposure, nvt (E 1 Mev)	Ultimate Burst Strength, psi	Elongation		Tube No.	Location*
		Uniform	Maximum		
	97,700	10	25	Control	-
1×10^{15}	86,200	7	9	6100	A
1×10^{16}	106,000	1	20	6061	A
1×10^{17}	97,000	§	17	5529	A
1×10^{19}	86,700**	-	-	6100	B
1.5×10^{20}	111,000	§	15	5529	B

* Location A. Section of tube bracketing area of contact with lower fuel element end bracket.
Location B. Section of tube immediately above Location A in higher flux region.

** This section failed when a small crack penetrated the wall. No elongation measurements were taken.

§ Less than measurable uniform elongation.

The burst tests were run to determine the effect of (1) the fretting corrosion, and (2) the total reactor environment on the room temperature burst properties. The corrosion marks that were observed in these tubes are rounded on the bottom, and are not sharp notches, but they have caused a definite wall thinning. This fact plus the potential for increased hydrogen content beneath the corrosion marks led to the assumption that the annealed section would be the weakest portion of the tube. The deepest corrosion marks on these tubes were located where the fuel element end brackets contacted the tube wall. The samples, from Location A, bracket these support marks and since this is at the end of the active portion of the fuel element there is a very steep gradient of fast neutron exposure (Figure 2). The exposure listed in Table II is that of the middle of the sample. The indicated scatter of burst strength of samples from Location A may be partly due to the difficulty in calculating the neutron exposure at this point. Samples from Location B were selected to determine the effect of total reactor environment on the burst properties. These samples were located in an area where the neutron flux was more uniform and the average exposure was higher. Two samples were taken from these areas. One of these was the only tube where a defect caused the failure at a lower stress than was expected.

The burst data to date are scattered, but there is an indication that the room temperature burst strength of the annealed Zircaloy-2 pressure tubes is increasing with exposure. Later results, soon to be published, confirm this strengthening with exposure.

Comparison of the burst test sections shown in Figures 4, 5, 6, 7, 8, and 9 reveals that both samples from Tube 6100 failed in an atypical manner because of the absence of a local bulge. The normal deformation pattern is a uniform increase in diameter along the length of the tube followed by a local bulging in the region where the fracture starts. Detailed examination of the test sections showed that the uniform expansion along the length of the sample decreased from about 10% elongation in diameter prior to irradiation to zero at the highest exposure. Disregarding samples from Tube 6100, the local bulging at the region of failure decreased from 25 to 15% with irradiation. There are insufficient results to date to be certain that these changes in deformation pattern at the point of failure are beyond the normal scatter of burst test results. The typical semiductile appearance of the fracture surface is shown in Figure 10.

In none of the three tests of sections from Location A did the presence of fretting corrosion marks from contact with lower fuel end brackets affect the failure characteristics of the tubes. However, a fretting corrosion mark from a bundle wire wrap in a section from Location B of Tube 6100 did initiate the tube failure. The lack of local bulging preceding fracture and the slightly lower burst strengths of the samples from Tube 6100 are indicative of failure initiated by a defect in the tube wall. Detailed examination of the section from Location A failed to detect any defect. Examination of the inner surface of the section from Location B revealed the fretting corrosion mark in the region of the crack shown in Figures 11 and 12. This mark was caused by contact between the pressure tube and the bundle wire wrap of a UO_2 fuel element. The corrosion mark is 3 mils deep and there is a 2-mil-deep layer of hydride under the mark. Apparently the deformation of the tube wall behind the hydrided area caused the many small cracks shown in Figure 12, most of which penetrate only through the hydride layer (Figure 13). As the stress increased several of these cracks began to propagate through the tube wall. Failure occurred when one of the cracks penetrated the wall. The crack was arrested even though the stress in the tube wall was at least 85% of the ultimate strength of the metal, which indicates that the tube behaved in a ductile manner. Theoretically, a short defect that propagates through a tube wall will end up with an effective crack length of twice the wall thickness at the time it penetrates the wall.(3) Crack arrest tests have been run on unirradiated tubing whose ductility was reduced by prior cold working by 50% reduction in area. The tests were run at room temperature. A crack twice the wall thickness in length was introduced into the tube wall while the tube was pressurized. When the pressure was equal to 80% of the tube ultimate strength, the crack propagated full length of the specimen.(4) When the pressure was 70% of the ultimate strength, the crack did not propagate. The irradiated sample from the annealed section exposed to 1×10^{19} nvt ($E > 1$ Mev), exhibits a greater fracture toughness than the unirradiated cold-worked tubing that was tested. The local yielding or wall thinning around the crack, as shown in Figure 5, is another indication of the degree of ductility that this tube has shown.

When the small crack popped through the tube wall, the pressure started to drop and the pump was turned off. The pressure within the tube decreased to 2500 psig at which time the leakage rate was less than could be measured with our present equipment. Apparently the local yielding around the crack at the high stress was sufficient to cause the crack to close when the stress was reduced by the drop in pressure. The 2500 psig that the tube maintained is greater than twice the operating pressure of the reactor.

Metallographic Examination of Wear Corrosion Marks

Although there have been a few instances of unexplained gouge-like marks on the internal surface of the pressure tubes, the most severe damage has been from the fretting corrosion marks formed at contact points between the pressure tube and fuel elements. Visual inspection in reactor has detected wear corrosion in all pressure tubes.(1) As these tubes were removed from the reactor, several of these wear corrosion areas were examined visually and metallographically.

Tube 6100. Defects observed in this tube were a small gouge just a above the lower fuel element support marks, top and bottom fuel element end bracket support marks, and spiral wire wrap marks from the bundle wrap. The gouge was 2 to 3 mils deep with a layer of cold-worked metal directly under the gouge (Figures 14 and 15). Top and bottom end bracket support marks ranged in depth from 2 to 3 mils for the top mark to 4 to 5 mils for the bottom mark. The area under the top mark contained a layer of massive hydride 2 to 3 mils deep while the area under the bottom mark was free from hydride that could be detected metallographically (Figures 16 through 22). One section of the tube cut for burst testing (Location B, Table II) contained a spiral wire wrap mark shown previously in Figures 11 and 12. Metallography of the defect revealed a layer of massive hydride about 2 mils deep directly under the corrosion mark (Figure 13).

Tube 6061. Fretting corrosion marks from the top and bottom fuel element end brackets were examined. For a 1-month operating period a Pu-Al element with wider end bracket support lugs was in this tube. These lugs were 1/4-inch wide versus a normal 1/16-inch width. The corrosion marks from the narrow lugs measured 3 mils deep at the top support mark and 6.5 mils deep at the bottom support mark. These marks occurred in a 2-week operating period and there was a slight increase in the hydrogen content under the top mark and very little, if any, increase under the lower mark. This hydrogen was in the form of platelets as compared to the granular type in Figure 18 (Figures 23 through 28). The maximum penetration of the wide lugs was less than 1/2 mil (Figure 29). However, the fuel element was removed and replaced at least once during the 1-month operating period; therefore, the maximum time any one lug could have been in contact with the tube wall was approximately 2 operating weeks.

Tube 5529. A lower fuel element end bracket support mark and an individual rod wire wrap mark near the lower support mark were examined. Both marks had the appearance of a contour relief map with three plateaus. The maximum depth of the support mark was 5 mils. A layer of massive hydride about 2 mils deep was found under the support mark (Figures 30 and 31). Only a slight increase in hydrogen was found under the wire wrap mark.

Crevice Corrosion of the Upper Flange

During the June 1961 Inspection of the pressure tubes, it was noted that all 85 tubes had areas of crevice corrosion where the stainless steel spiral-wound, asbestos-filled gaskets contact the top portion of the flange. These areas varied from small patches to a 1/16-inch-wide ring around the entire top of the flange.

Figure 32 shows a typical cross section of a flange. This is a screwed flange with two seal welds. The top weld has a minimum of 0.020-inch penetration and is in contact with the primary coolant. Excessive corrosion at the top seal weld could penetrate into the threads and possibly cause a blow-out of the lower seal weld.

Two pressure tests were run on unirradiated flanges that were machined to allow pressure to be applied to the threads. These flanges were held in a fixture similar to the in-reactor connecting joint. The lower seal weld started to leak at 2200 psig at 550 F indicating a factor of 2+ over operating pressure.

The flanges from tubes numbered 6100, 6061, and 5529 were destructively examined. The corroded areas from the flange from Tube 6100 was 2 to 4 mils deep with 3 to 4 mils of massive hydride directly under the corroded area (Figures 33 and 34). Several areas of porosity were noted in the seal weld on the top surface of the flange. One of these holes had corrosion product in it and an associated layer of hydrides (Figures 34 and 35). Similar areas were examined from Tubes 6061 and 5529, however, no hydrides were found beneath corroded areas. From the examination of these three flanges, the extent of corrosion in these areas does not appear to be increasing at a measurable rate.

ACKNOWLEDGEMENTS

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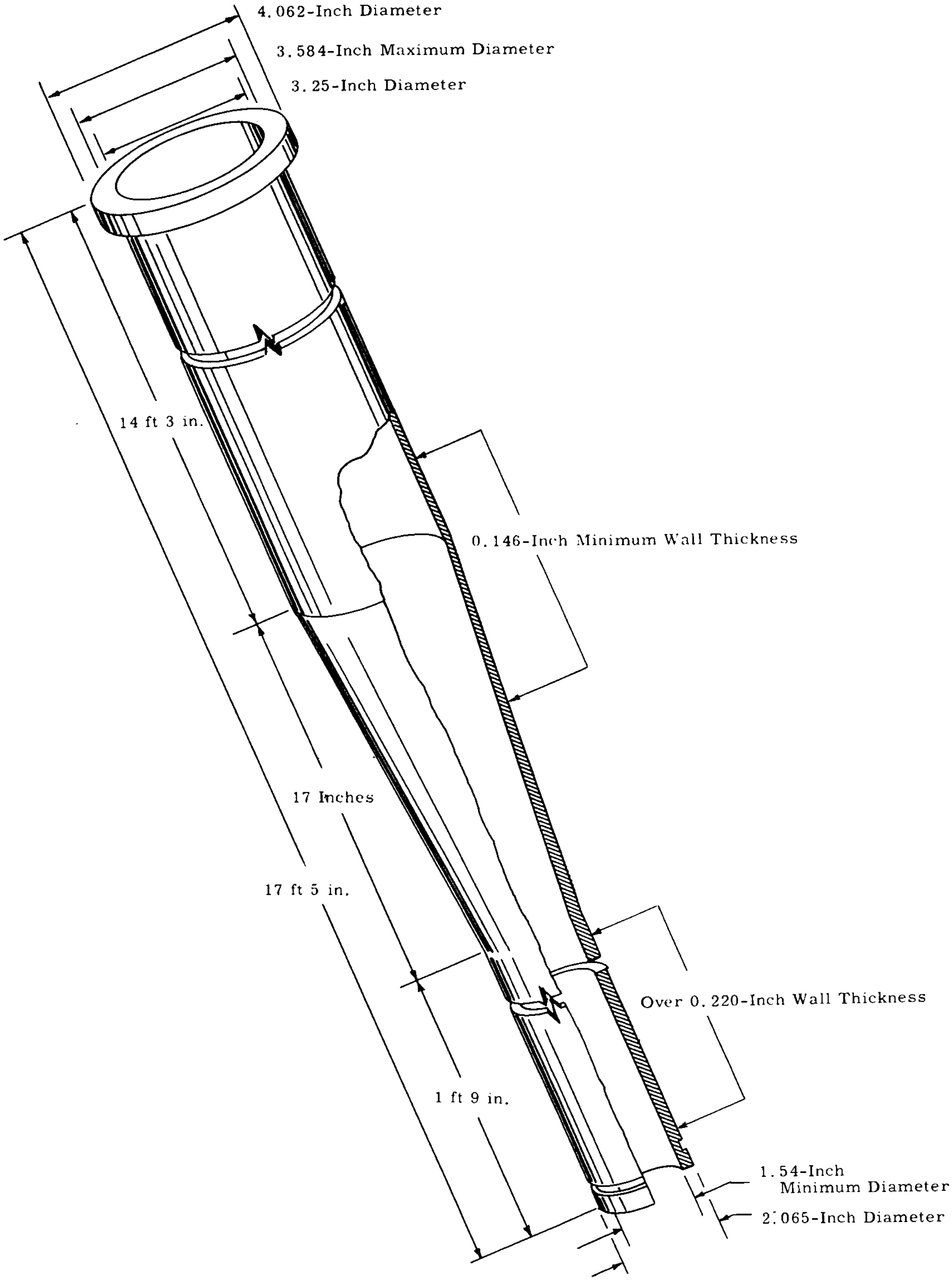

FIGURE 1

PRTR Process Tube with Nominal Dimensions

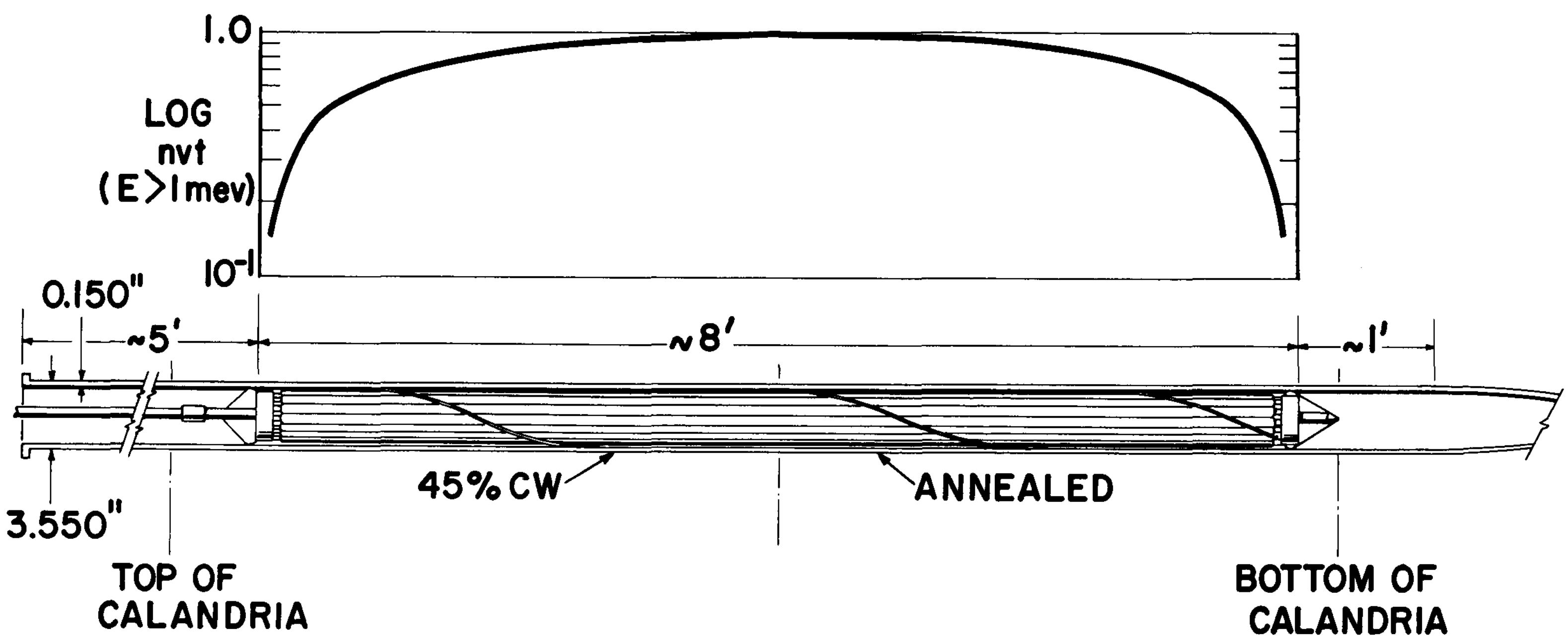

FIGURE 2

PRTR Pressure Tube Assembly Showing Position of Fuel Element in the Tube Relative to Fast Neutron Exposure over the Active Portion of the Element, and the Approximate Location of the Annealed and Cold-Worked Portions of the Pressure Tube

Neg. No. 0621842-1
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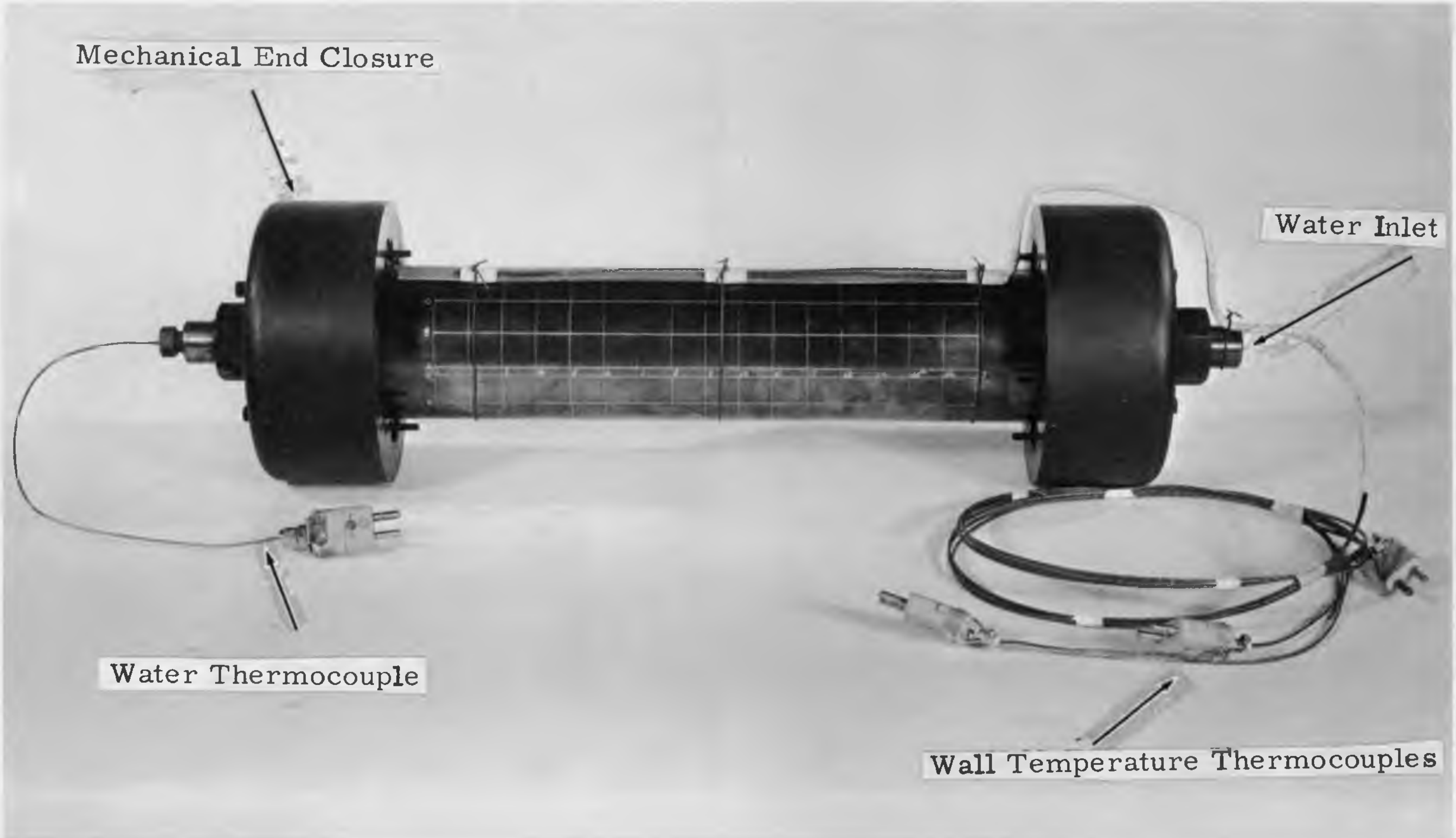


FIGURE 3

Burst Test Assembly
with Mechanical End Closures and Thermocouples Installed
(Grid is 2 cm square.)

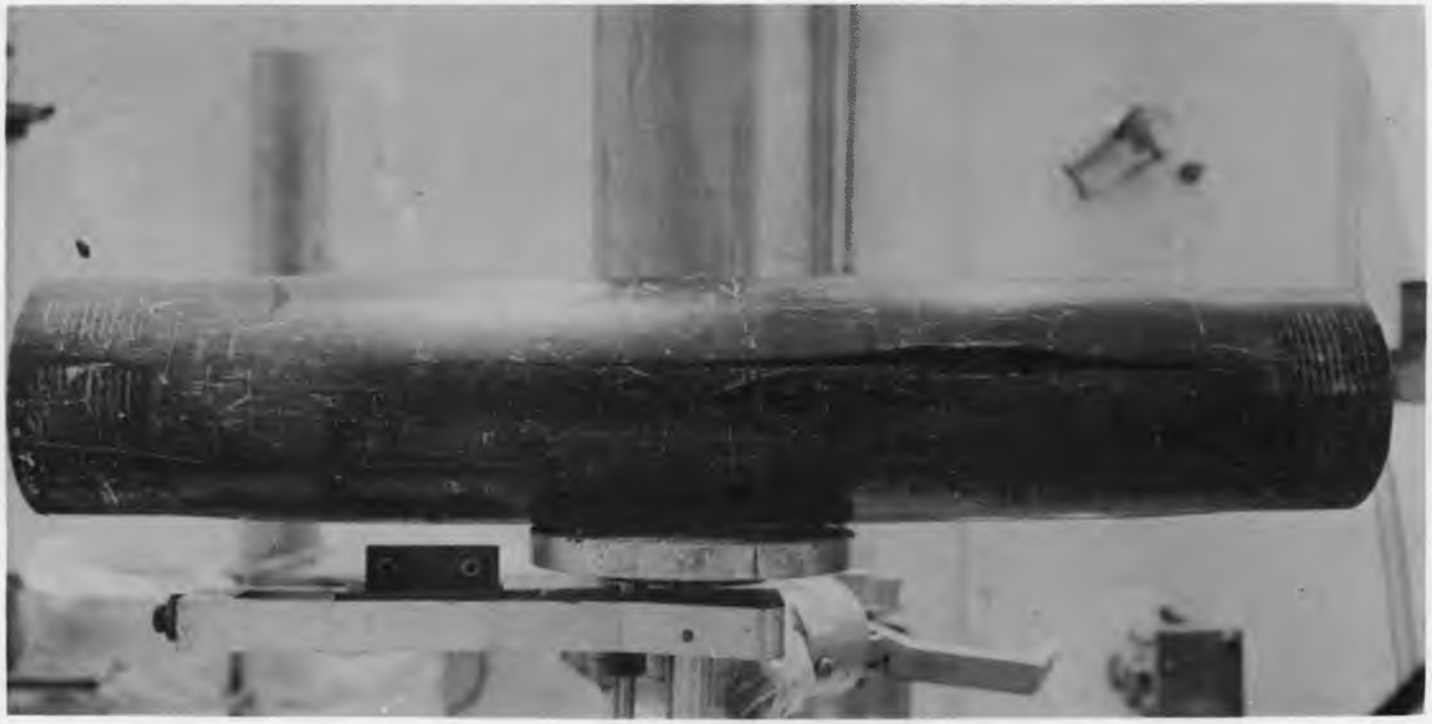


FIGURE 4

Room Temperature Burst Test Sample from Pressure Tube 6100, Location A, Removed After 2 Weeks Operation. 0.35X

FIGURE 5

Room Temperature Burst Test of Tube 6100, Location B, Immediately Above the Sample in Figure 4
(Failure occurred when a crack that started in a wear corrosion mark propagated through the tube wall. The crack is about 3/16-inch long on the outer surface of the tube.)

FIGURE 6

Room Temperature Burst Test Sample from Pressure Tube Number 6061, Location A, Removed After 1-1/2 Months of Operation. 1/2X

FIGURE 7

Room Temperature Burst Test Sample from Pressure Tube Number 5529, Location A, Removed After 2-1/2 Months of Operation. 1/2X

FIGURE 8

Room Temperature Burst Test Sample from Tube 5529, Location B,
Immediately Above the Sample in Figure 7
(This sample had the highest accumulated fast neutron exposure of the burst test sections.) 1/2X

FIGURE 9

Close-Up of Fracture from Burst Sample in Figure 6
(This tube appeared the least ductile of any sections tested.) 1X

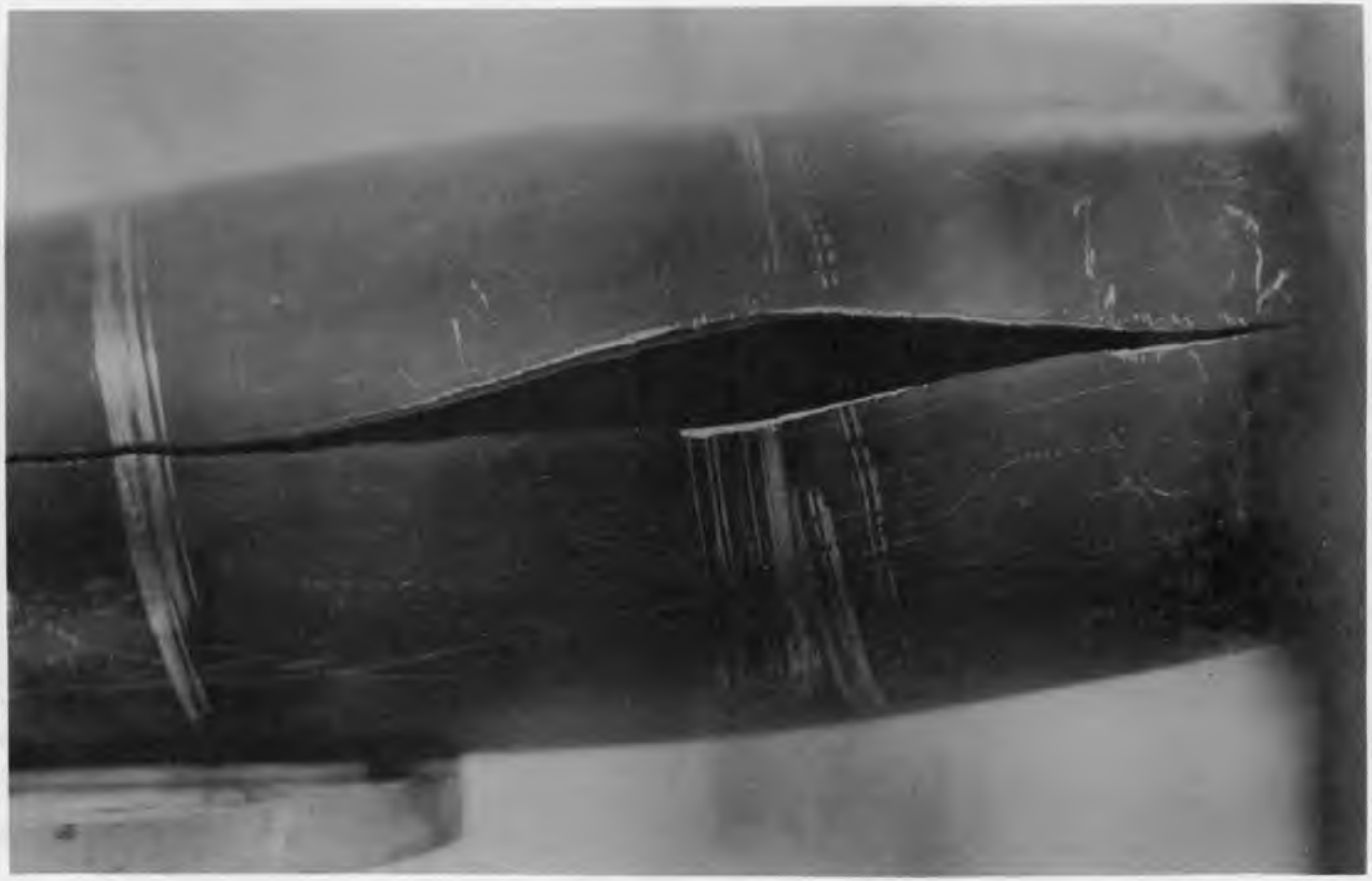


FIGURE 10

Close-Up View of Fracture Area from Figure 8
(Note shear area on inner and outer surface near point of failure, estimated to be 30% of wall thickness.)

FIGURE 11

Wear Corrosion Mark on Inner Surface
of Tube Caused by Bundle Wrap
(Sample is from pressure Tube 6100 shown in Figure 5.) 2X

FIGURE 12

Same Area Shown in Figure 11
(Large cracks continue through the wall. Smaller
cracks extend the depth of the 2-mil-deep hydride layer
under the corrosion mark.) 4X

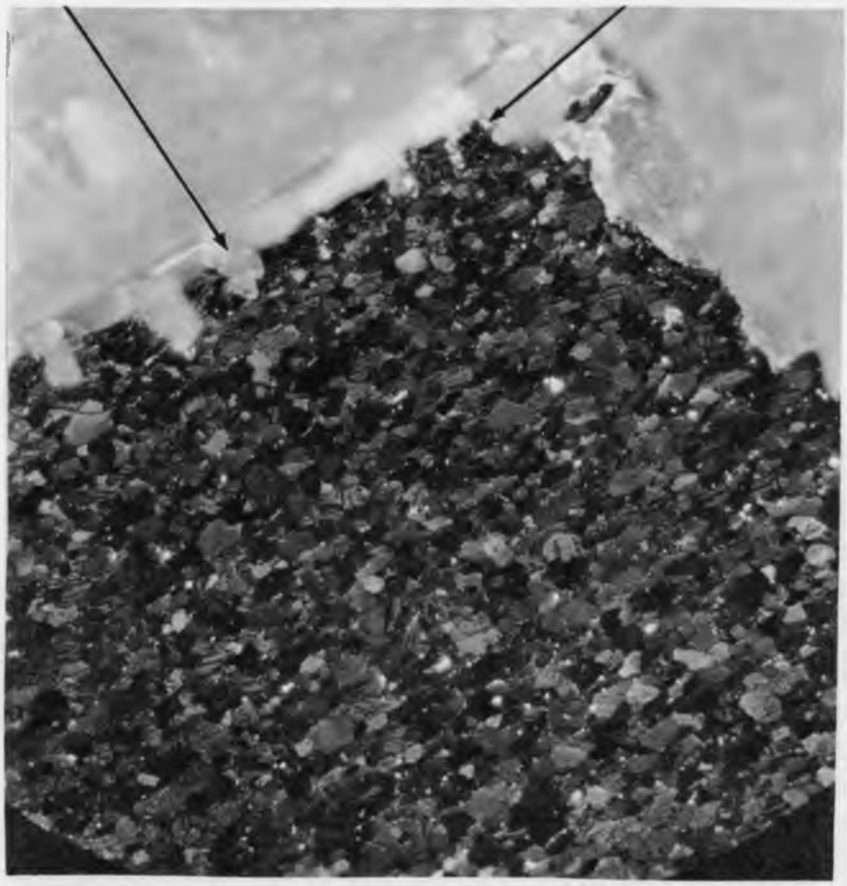


FIGURE 13

Traverse Section of Area in Figures 11 and 12
Showing the Cracks Extending Through the Layer of Massive Hydride
(This layer is 1-mil deep in this area.) 250 X

FIGURE 14

Replica Made of Gouged Area
in the Tube Wall of Process Tube 6100
(Maximum depth, 2-3 mils) 3X

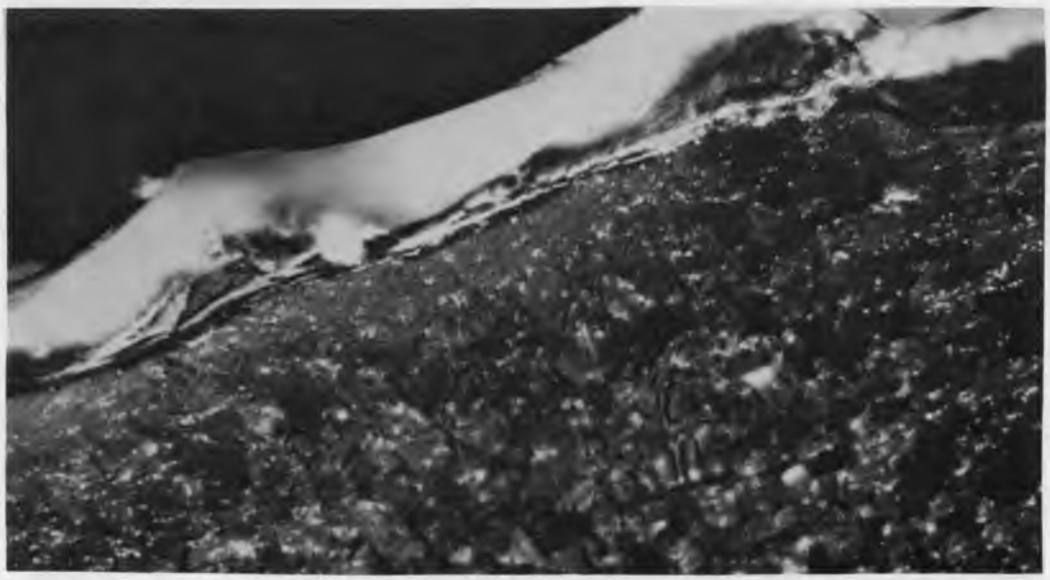


FIGURE 15

Grain Structure Under Gouge Shown in Figure 14
(Cold-worked structure at surface is 1-mil deep.) 500X

FIGURE 16

Process Tube 6100 Wear Corrosion Marks
with Individual Rod Wire Mark on Left
(Two distinct wear areas show the fuel element shifted during operation. Depth is 2 mils. Upper fuel element support mark is on the right. Note white corrosion product.)

FIGURE 17

Longitudinal Section Through Upper Fuel Support Mark,
B Etched and Anodized
(Area near the top with no grain structure is layer of hydrides.)
Polarized Light, 250X

FIGURE 18

Same Area as Figure 17
(Layer near the top of picture
shows massive hydrides. Depth is 2 mils.)
Bright Field, 250X

FIGURE 19

Replica of Lower Fuel Element Support Mark on Process Tube 6100
(Depth is 4-5 mils.) 5X

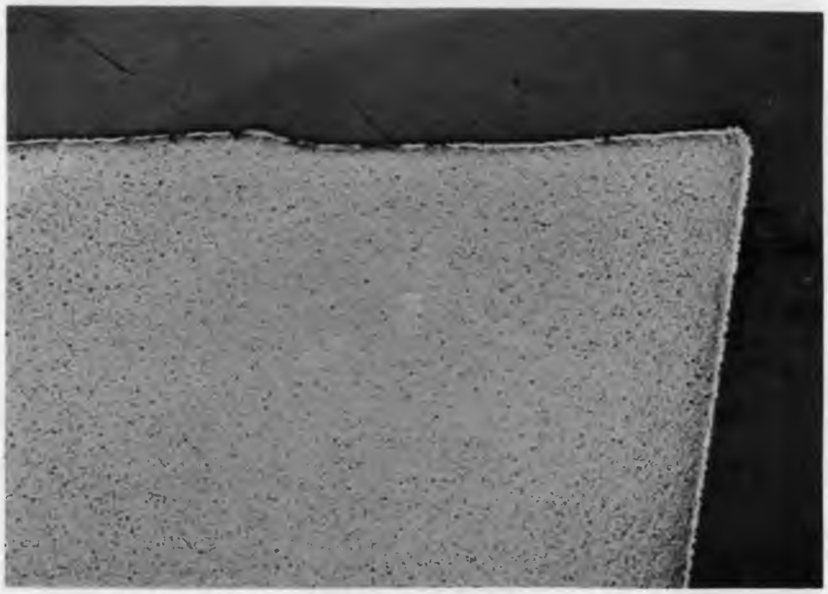


FIGURE 20

Transverse Section Through Localized Corrosion Area
at Lower Fuel End Bracket Support Mark from Figure 19
(Maximum depth at this point is 2-3 mils.)
Bright Field, 50X

FIGURE 21

B Etched and Anodized Edge
of Corrosion Area from Figure 20
(Compare with Figure 17.)
Polarized Light, 250X

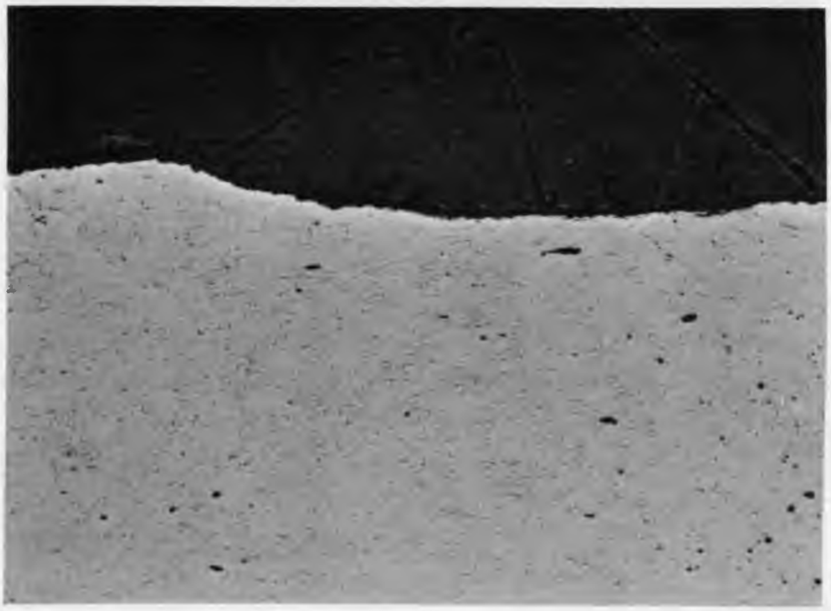


FIGURE 22

Same Area as Figure 21
(No hydrides are visible in this area.)
Bright Field, 250X

FIGURE 23

Process Tube 6061 Wear Corrosion Area
from the Upper Fuel Element Supports
(Maximum depth is 3 mils.)

FIGURE 24

Transverse Section Through Upper Fuel Support Mark
Shown in Figure 23
(Typical Cold-Worked Structure. B Etched)
Polarized Light, 250 X

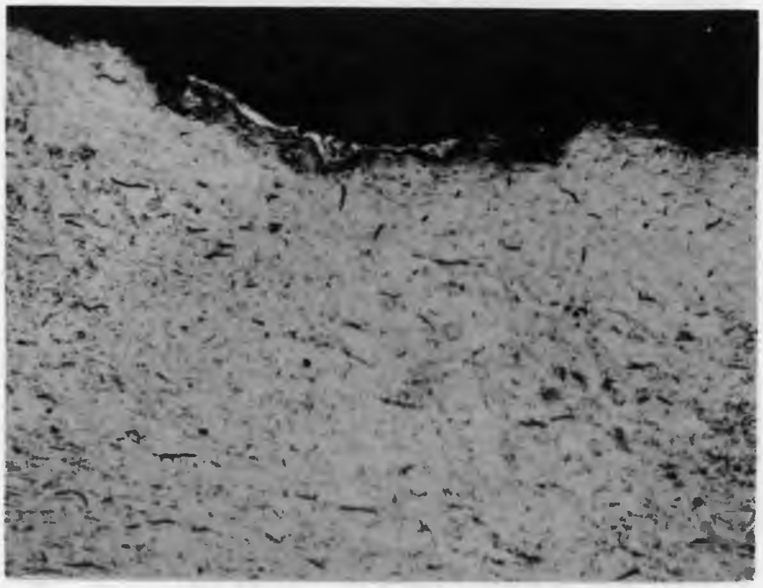
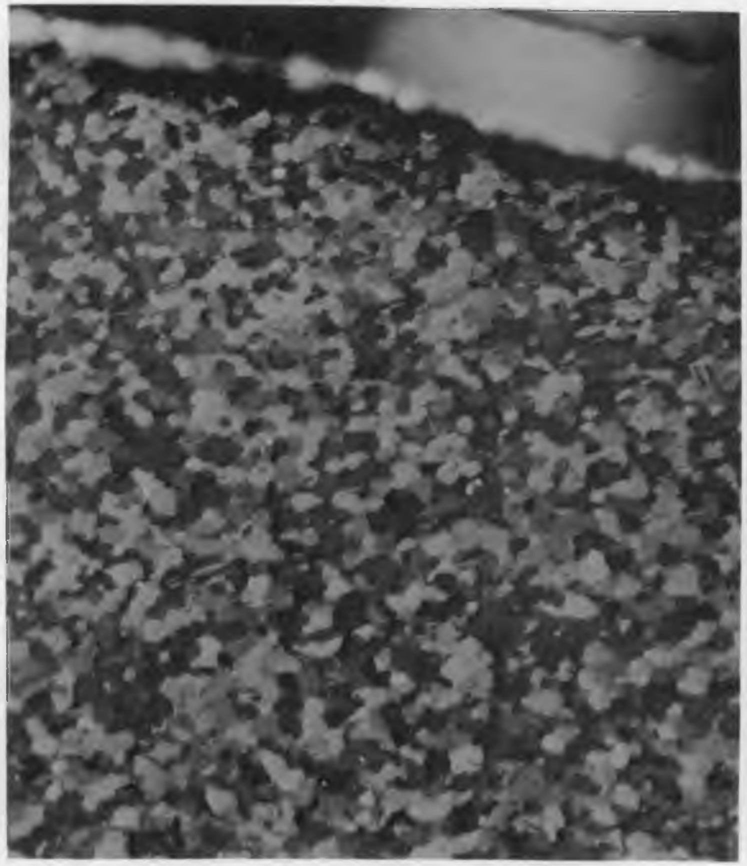


FIGURE 25

Same Area as Figure 24
(Note the slight increase in hydrides under corrosion area.)
Bright Field, 250X

FIGURE 26

Process Tube 6061 Lower Fuel Element Support Mark
(The corrosion mark in the center of the sample is 6.5 mils deep. This occurred during the first two weeks of operation and was caused by the 1/16-inch wide supports.) 4X



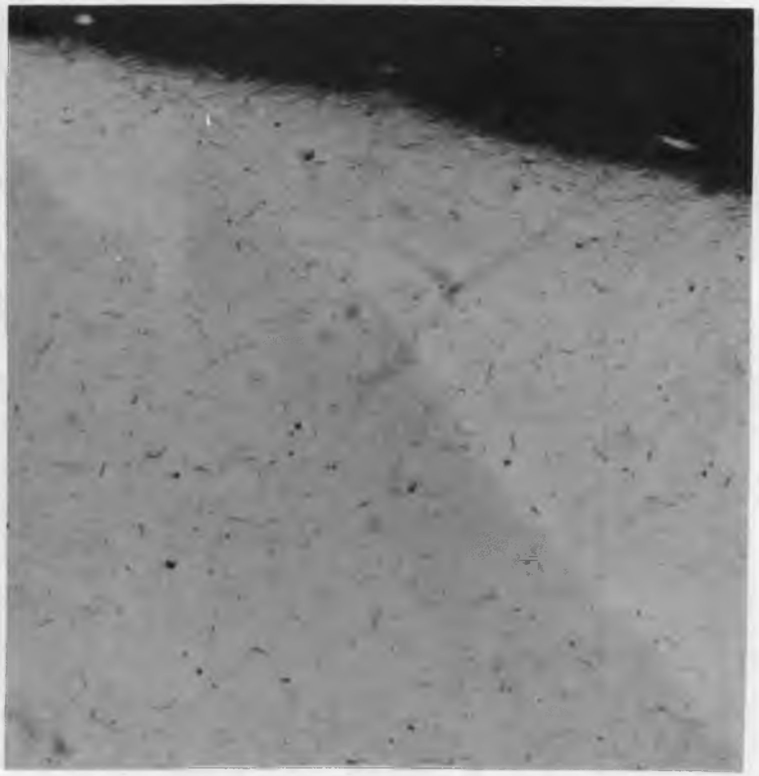


FIGURE 27

Transverse Section of the Shallow End
of the Corrosion Mark from Figure 26
(Typical Annealed Structure
Found in the Lower Half
of the Pressure Tube. B Etched)
Polarized Light, 250X

FIGURE 28

Same Area as Figure 27
(Very Few Hydride Platelets
Under Corrosion Area. B Etched)
Bright Field, 250X

FIGURE 29

Typical Wear Corrosion Marks on Process Tube 6061
from 1/4-Inch-Wide Fuel Supports
(One Pu-Al element was equipped with special 1/4-inch-wide supports
to determine the effect on wear corrosion. The maximum penetration
that occurred was less than 1/2 mil during a two-week operating period.)
5X

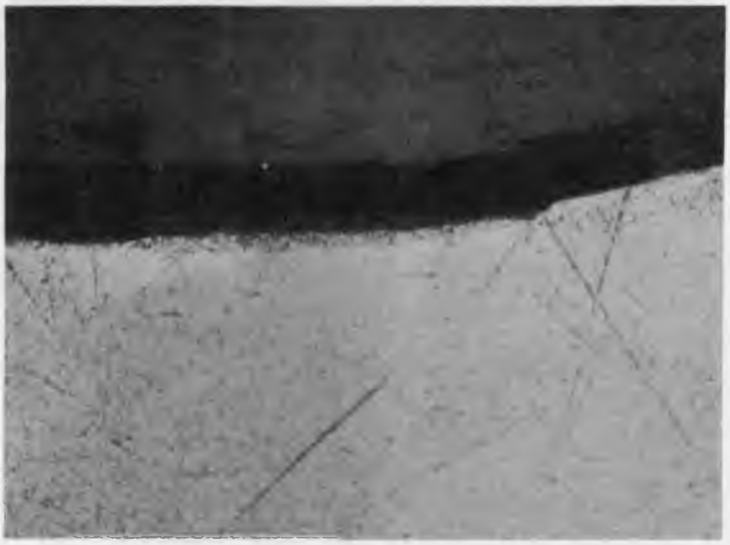


FIGURE 30

Wear Corrosion Mark
from Lower Fuel Element Support in Tube 5529
(Maximum depth was 5 mils.) 50X

FIGURE 31

Same Area as Above
(Layer of massive hydrides is 2-mils deep. B Etched)
Bright Field, 250X

Top Seal Weld
Corrosion
Noted in this Area

Bottom Seal Weld

FIGURE 32

Top Flange Construction Showing Threads and Location of Seal Welds.

FIGURE 33

Top Flange from Tube 6100
(Heavy white ring is corrosion product. Light rings are places where the gasket has cut through the oxide film.) ~2X

FIGURE 34

One Corrosion Area from Figure 33, Showing Gray Layers of Oxide (Upper Left) and Gas Pocket in Seal Weld with Gray Layer of Oxide at Lower Right in the Hole. As Polished, 100X

FIGURE 35

Same as Figure 34
(Note massive hydrides in beta phase structure beneath oxide at upper left and at lower right in the hole. B Etched)
Bright Field, 250 X

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