NMI-4364

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Interim Summary Report

Corrosion Behavior of Defected Fuel Elements with U - 2 $^{w}/_{o}$ Zr Core Clad with Zircaloy-2

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July 21, 1958

Nuclear Metals, Inc.
Cambridge, Massachusetts

Contract No. AT(30-1)-1565
Sponsor Agreement No. S-31

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ABSTRACT

The aqueous corrosion behavior of defected fuel elements having a U - 2 w/o Zr core with Zircaloy-2 cladding has been studied in 212° to 660°F water. A standard diffusion heat treatment for 7 hours at 880°C, followed by moderately rapid cooling, was established to overcome the effect of subtle defects in 15-mil Zircaloy cladding. The development of this heat treatment was carried out primarily with small rod specimens, which were also used to obtain data on auxiliary effects of the diffusion heat treatment. The heat treatment has been applied to full diameter tubes. One of these tubes was tested with subtle defects and various sections have been observed following the insertion of gross defects. These observations have shown a difference in the rate of hydrogen evolution as a result of heat treatment. In addition, the heat treatment changes the nature of failure in both full diameter tube sections and small rods.

The presence of 2 w/o Zr in the core reduces the corrosion rate considerably. Comparative quantitative data are presented for ~~uranium~~ U alloys with various ~~zirconium~~ Zr contents up to 15 w/o, in 212° to 650°F water. Zircaloy-2 was tested in 750°F steam.

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I. INTRODUCTION

Corrosion behavior is a prime consideration in the design of a fuel element to be used in a water-cooled reactor. For the current program, a tubular fuel element was specified with an OD of 2.060 inch and an ID of 1.467 inch. Both the inner and outer Zircaloy-2 cladding have a nominal thickness of 15 mils. The core alloys is U - 2 $^{w}/_{o}$ Zr. Since exposure of such a high uranium alloy to high temperature water can lead to serious consequences, it is necessary to achieve maximum assurance of the integrity of the cladding and the end seals. Methods of producing reliable cladding by coextrusion have been developed in this laboratory.[1] In addition, an interdiffusion heat treatment was developed to overcome inadequacies in the cladding of small diameter rods.[2,3,4] A standard heat treatment has been developed and applied to full diameter tubes. This report includes data on the rate and nature of failure of full diameter tubes resulting from intentionally inserted cylindrical defects in the cladding. Such data indicate whether a failure will be detected early enough to permit removal of a fuel element before either serious mechanical damage can occur or excessive uranium enters the coolant as a result of corrosion

The effect of addition of zirconium to the core has been determined with clad samples with either drilled defects or with the flat ends of transverse sections exposed. Such samples with flat ends exposed are also being used to advance the understanding of the mechanism of corrosion.

II. INTERDIFFUSION HEAT TREATMENT

The purpose of the interdiffusion heat treatment is to overcome the effect of inadequacies in the cladding which might not be detected in routine inspection. Almost all the information on the application of the heat treatment for this purpose has been obtained with short specimens (about 2 inches), having a diameter near 3/8 inch. A "subtle" cylindrical defect is used, having a diameter of 7 mils and stopping at the core. Before this defect is inserted, the section of rod has been bored out at the ends and appropriate Zircaloy-2 end plugs inserted and Heliarc welded. In standard practice, these welds are checked by an overnight test in 650°F water.

The results of the earlier work are first summarized and an explanation suggested for the effectiveness of the heat treatment. Data are then presented on various rod samples having the 15-mil clad thickness which has been adopted for the full diameter tubes. These data provide the basis for the choice of the standard heat treatment, applied to the tubes. Results are then given for a short, full diameter tube which was heat treated, checked for clad integrity, provided with 7-mil defects, and corrosion tested. Various considerations related to the heat treatment are also discussed. Unless otherwise indicated, the core composition remained fixed as the U - 2 $^{w}/o$ Zr alloy.

A. Earlier Work

Lamartin and Levine[2,3] established that a suitable interdiffusion heat treatment overcame the effect of a 7-mil defect which would otherwise have led to rapid failure as a result of exposure of the U - 2 $^{w}/o$ Zr core to high temperature water. Even though the defect inserted before or after heat treatment exposed some of the core, the heat treatment had a protective effect. Corrosion did start but was prevented from spreading. The net effect, even after long term exposure, was compaction of a small pocket of corrosion product with very little if any deformation of the Zircaloy cladding, even around the defect. Jenkins[4] continued and extended this work, investigating the extent to which the effectiveness of the heat treatment was affected by factors such as defect diameter and depth, clad thickness, bond strength, duration and temperature of heat treatment. Thus, it was clearly established that there is no unique range of temperature for effective heat treatment. Various temperatures can be used if sufficient time (e.g. 2 hours at 1050°C) is allowed for the interdiffusion necessary to provide protection. The necessary interdiffusion depends on such factors as clad thickness and defect diameter.

This work provided several lines of evidence, outlined below, that the effectiveness of interdiffusion is due to its strengthening of the bond between the core and the cladding. (It is worth noting that uranium and zirconium are compatible. Since no brittle compounds are formed, core-cladding interaction is likely to be beneficial.)

1. The effectiveness of the heat treatment is not due to the formation of a corrosion-resistant layer. Protection is found even when the defect exposes core material beyond the diffusion layer. Sections of corroded specimens leave no doubt that corrosion started early in the period of test.

2. The effectiveness of the heat treatment does depend on the initial presence of a good core-cladding bond. If the bond in the fabricated composite is inadequate, a defected sample will fail if given the heat treatment suitable for a well bonded composite. It may be possible to overcome the inadequacy of the fabricated bond by a more severe heat treatment.

3. Bond quality, as shown by mechanical tests, is improved by a diffusion anneal when the core is U - 2 w/o Zr. With other cores, such as unalloyed uranium or thorium, a diffusion anneal does not improve the bond strength or the corrosion resistance.

4. The severity of the diffusion anneal necessary to overcome the effect of a defect increases as the clad thickness decreases. Thinner cladding is less capable of resisting stress due to expansion of uranium on conversion to oxide, and therefore requires greater strengthening of the

core-cladding bond. A minimum clad thickness, perhaps 10 mils, may be a necessary condition for even the most severe heat treatment to overcome the effect of a defect. Thicker cladding leads to survival after heat treatment even if defects penetrate more deeply into the core.

B. Selection of Heat Treatment for 15-Mil Cladding

After the cladding thickness had been set at 15 mils, a series of experiments was undertaken with 3/8-inch rod samples to establish the length of heat treatment necessary at various temperatures to assure protection against 7-mil defects penetrating to the core. To place a more severe demand on the heat treatment, each sample was provided with four defects around a circumference at the center of the sample. (It was subsequently found that corrosion tests of samples with "borderline" heat treatments led to failure at only one of the defects.)

The defects were inserted before heat treatment by an "automatic" method developed by R. G. Jenkins. A weight is affixed to the spindle arm of the small drill press, so that a constant load drives the drill through the metal. A dial indicator shows the rate of penetration of the drill into the metal. This rate is steady as the drill bores out Zircaloy, but there is an abrupt decrease in speed as the drill enters the uranium alloy core. The observer is guided of course by prior knowledge of the approximate clad thickness. The depth of penetration of this or any other defect is checked with a toolmaker's microscope to which is attached a dial indicator. It is then possible to measure the motion of the column when the change is made from focusing on the rim of the defect to its base. The success of the Jenkins method of inserting defects is shown by a section through the defect (Fig. 1) as well as by the failure of samples which received inadequate heat treatments.

The earlier work had emphasized heat treatment above 1000°C. Since large scale operation was expected to be facilitated at lower temperatures, it was decided to establish the minimum heat treatment times for lower temperature. The 800 to 900°C range seemed reasonable. Early experiments eliminated the lower portion of this range because of the coarsening of the Zircaloy grains. Attention was then concentrated on 900 and 880°C, the latter being near the maximum operating temperature of an industrial salt bath whose use was then contemplated for heat treatment of full size tubes.

The results of the corrosion tests are summarized in Table I. The test medium was 650°F (343°C) water. Tests were arbitrarily terminated after an accumulated test period exceeding 1000 hours. On the basis of these tests, it was concluded that a heat treatment for 7 hours at 880°C or for 5 hours at 900°C was adequate to overcome the effects of 7-mil defects to the core. These two heat treatments are not necessarily equivalent since at each temperature, slightly shorter heat treatments might be adequate.

C. Tests of Sections from Full-Diameter Tube with 7-Mil Defects after Heat Treatment

Other considerations, including the limited stock of full diameter tubes (2.060 OD x 1.467 ID), prevented the testing of such stock in a manner similar to that used for the small rod specimens discussed above, i.e. insertion of defects before heat treatment. A short, full diameter tube with integral end seals was available, however, after it had been heat treated for 7 hours at 880°C, and then shown to have sound cladding by a 24 hour test in 750°F (399°C) steam. Four 7-mil holes were then inserted around the outer circumference of this tube. The Jenkins "automatic" drilling method was used but uncertainty was introduced by the diffusion band produced by the heat treatment. Microscope measurements showed three of these holes to have depths of 17 mils while one measured 16 mils. (Subsequent metallographic examination of a section near these holes showed that the clad thickness after heat treatment was 13 mils and the width of the diffusion band, 4 mils.) The tube was then tested for a total of 28 hours in 600°F (316°C) water and no swelling of the cladding was seen around the defects.

A second, deeper set of four 7-mil defects was then inserted about 1/2 inch away from the first set. Three of these defects were found to be 20 mils deep; the fourth was closer to 19 mils. In a four hour test in 600°F (316°C) water, serious opening and distortion of the cladding occurred around two of these holes, but swelling was very slight around the other two, especially the one which had measured about 19 mils. During this additional 4 hour test, the first set of more shallow holes appeared unchanged. A section through one of these holes (Fig. 2) clearly established that this set of holes was deep enough to permit exposure of the core during the test. Thus, the shallow holes to the core were prevented by the heat treatment from causing rapid failure. The deeper holes into the core did lead to failure in spite of 7 hour-880°C heat treatment for nominal 15-mil cladding.

Paralleling these tests with the full diameter tube, similar tests were performed with two pairs of 3/8-inch rod samples, defected after heat treatment. One pair had four 7-mil defects measuring 16-mils deep and presumed to have reached the core. This pair survived over 1000 hours in 650°F (343°C) water. In the other pair, the defects were 20-mils deep. One of these samples also survived over 1000 hours in test. The other one suffered failure, apparently at only one defect, between 210 and 261 hours in the 650°F water. Again, the effectiveness of the heat treatment depends on the depth of the defect. In spite of the difference in time to failure for deeper defects, these results indicate that the heat treatment is likely to overcome shallow defects in large tubes as in small rods.

D. Effect of Cooling Rate

The thermal history after the diffusion anneal may undo the beneficial effect of the diffusion anneal. Previous work indicated that between 750 and 550°C, the beneficial effect may be lost. The possibility of such a harmful effect has to be considered in connection with two important points. First, steps have to be taken to assure as rapid a cooling rate following the heat treatment of full size tubes as is obtained in the so-called air cooling of the rod specimen. (In the air cooling, it is the sealed Vycor tube which is really air cooled, the specimen being left in the tube until both are near room temperature.) Second, a supplementary heat treatment at a temperature below the diffusion anneal temperature may be recommended for anisotropic dimensional stability under irradiation. In fact, it was during investigation of the effect of such supplementary heat treatments that Jenkins discovered the harmful effect which may occur at temperatures below the diffusion anneal temperature.

Several modifications of cooling were used to demonstrate this effect and define the critical temperature range:

1. The sample is furnace cooled from the diffusion anneal temperature to 650 or 550°C and then air cooled;

2. After air cooling, samples are maintained at a fixed temperature (600°C) for different lengths of time and then air cooled again;

3. Samples are transferred from the diffusion anneal furnace to a furnace at a lower fixed temperature (690°C), where they are held for different lengths of time and then air cooled.

Table II gives the results of such modifications of cooling after the standard 7 hour - 800°C heat treatment for rods with nominal 15-mil cladding. The various modifications of the cooling show the existence of a sensitive range in the neighborhood of 600°C. The time necessary to obtain a harmful effect at 600° is shorter than at 690°C. With proper cooling, the time spent in the sensitive range can be made short enough to avoid jeopardizing the benefits of the anneal. Following the heat treatment of full diameter tubes, the evacuated steel container is quenched in water, giving a cooling rate very similar to that of rod specimens when the Vycor container is air cooled. Confirmation of the adequacy of this cooling following a large scale heat treatment is obtained with the help of small defected rod specimens. These specimens, held in contact with a large tube during the heat treatment, subsequently survived over 1000 hours in the 650°F water test.

The cause of this sensitive range is thus far unexplained.

E. Effect of Irradiation

As in other metallurgical treatments, consideration must be given to the possibility that the beneficial effect of the diffusion anneal may be undone by irradiation. Only limited data are available regarding this point, but they are not at all discouraging. Defected samples given the then standard 2 hour - 1050°C heat treatment were irradiated by Argonne National Laboratory in the MTR to a burn-up of about 0.1 a/o. Subsequent corrosion tests showed that the protective effect of the diffusion anneal had not been impaired by the irradiation.[5] Additional samples have been submitted to Argonne for higher burn-up.

F. Auxiliary Effects

Several possible unfavorable effects of the diffusion anneal have to be considered: diffusion of uranium into the Zircaloy cladding, extending so far as to produce an excessive concentration of uranium at the surface; impairment of the Zircaloy's corrosion resistance; and excessive coarsening of the Zircaloy's grains. Data given in the appendices show that diffusion anneals even more severe than the standard 7 hour - 880°C treatment do not produce any of the unfavorable effects.

III. EFFECT OF VARYING ZIRCONIUM ADDITIONS TO CORE ALLOY

Additions of zirconium improve the corrosion resistance of the uranium core. The effect of varying zirconium additions has been established in two ways. In one way, corrosion rates of the bare alloy are measured, and in the second, the behavior of coextruded rods with intentional defects is studied. These comparisons were made with four different types of defects whereas other tests covered in this report involved either a "subtle" 7-mil defect (SectionII) or a gross 40-mil defect with total depth of 40 mils (Section IV).

A. Bare Alloys

Transverse sections of coextruded rods (or tubes) are conveniently used for the determination of corrosion rates. Since the core is exposed only at the flat ends, the exposed area remains constant as long as the cladding remains undamaged. Such sections have been used to obtain corrosion rates at several temperatures for alloys with various zirconium contents. Table III summarizes these data and compares them with rates reported by other laboratories. Although this method of measurement may introduce uncertainty in the absolute values of the rates, these data are particularly valuable for comparing different alloys at the same temperature. It is quite clear that at any temperature from 100°C to about 300°C, the addition of 2 w/o Zr reduces the corrosion rate by a factor of about 3. Further additions of zirconium have a much smaller effect than the first 2 w/o.

Any effect of the diffusion heat treatment on the 2 w/o Zr alloy is so slight that even the direction of the effect is difficult to determine.

B. Defected Samples

The beneficial effect of the addition of 2 w/o Zr to the core is shown by comparison of samples which have been defected and tested in a similar manner, but differ in core composition. All the samples shown in Fig. 3 were tested in the as-extruded condition. Set A, on the left, had an unalloyed uranium core, while Set B, on the right, had a nominal U - 2 ^{w}/o Zr core. Samples were tested in 440°F (227°C) water with four types of drilled defects: diameter, 7 or 40 mils; depth 25 or 40 mils. The accumulated duration of test for each specimen is shown in the tabular legend accompanying the photograph. The samples with the alloyed core are in better condition after 15 hours than those with the unalloyed core after 3 hours.

Only a limited number of experiments were performed with rods whose core contained more than 2 ^{w}/o Zr. These experiments indicated that even up to 10 ^{w}/o Zr, the additional zirconium had only a slight effect on the corrosion behavior.

IV. PROGRESS OF CORROSION: EFFECT OF HEAT TREATMENT

Results on the progress of corrosion resulting from an intentional defect are available in several forms. Visual data on short rod samples provide a qualitative comparison showing the benefit of the diffusion heat treatment in reducing the magnitude and also the pattern of failure. Annealed rod samples were also used to follow the progress of corrosion at two different temperatures. Quantitative data on the progress of corrosion were obtained with sections from full-diameter tubes.

A. Comparison of Short Rod Samples

The effect of the diffusion anneal is seen in the samples shown in Fig. 4, which is arranged similarly to Fig. 3; the as-extruded Set C on the left is compared with the heat treated Set D on the right. The test temperature is now 530°F (277°C). The lengths of the tests were more nearly equal than they were for the comparison shown in Fig. 3. Nevertheless, the beneficial effect of the heat treatment is seen even with 40-mil diameter defects. In addition to reducing the severity of the failure, the heat treatment modifies the nature of the failure. In as-extruded material, the cladding bulges and cracks unevenly and failure progresses longitudinally. Heat treatment seems to localize the failure, which develops with radial symmetry around the defect.

Similar differences as a result of heat treatment were also seen in shorter tests at 572°F (300°C).

These differences in the pattern of corrosion can be attributed either to metallurgical changes in the core and/or cladding, or to the interdiffusion. If the main effect of the heat treatment is modification of the core or the cladding (due to the material's entering a new phase region), shorter heat treatments would be expected to produce the same results as the standard interdiffusion treatment. Figure 5 shows the appearance of four sets of samples tested for 4 hours in 530°F (277°C) water after different heat treatments. From these results, it is inferred that the change in the pattern of failure resulting from the 7 hour -

880°C treatment is due not only to the temperature but to the duration of the heat treatment, and hence is attributable to interdiffusion, which is a function of time.

All these experiments on the effect of heat treatment were performed with samples containing 2 w/o Zr in the core. With unalloyed uranium, very little if any improvement in corrosion behavior was derived from the heat treatment. A heat treated set would strongly resemble the as-extruded Set A (Fig. 3). This difference in the response of unalloyed uranium has not been intensively studied. Presumably, the Zircaloy cladding can extract carbon from the unalloyed core and form a zirconium carbide layer [6,7] which weakens the band. With the unalloyed core, heat treatment does not improve the bond and may even worsen it.

B. Progress of Failure in Small Rod Specimens

The stages in the progress of failure generated by a defect can be established by observing samples which have been in corrosion for different lengths of time. Rod samples with 15-mil cladding were provided with defects whose diameter and depth were both 40 mils. After the standard diffusion anneal, a set of 3 samples was run together for the same time at a fixed temperature. Sets run for different lengths of time could then be mounted to show the stages in the progress of corrosion. Presumably, this sequence would be seen in a single sample if it could be observed continually. This method of using separate samples for the different intervals reduces uncertainty introduced by corrosion during heating and cooling periods. The importance of behavior during relatively slow heating to fairly high temperature is seen in the "0" hour set at 570°F (299°C) shown at the top of Fig. 8. This set was merely heated to the test temperature and cooled without spending any time at temperature.

Equivalents of time lapse photographs, obtained with samples tested for uninterrupted periods, are shown in Figs. 6 and 8 for 530°F (277°C) and 570°F (299°C) respectively. The 530°F samples readily lent themselves to sectioning and polishing right up to the center of the defect, so that the penetration of corrosion into the core could be observed, as shown in Fig. 7. At this temperature, failure is quite slow. The interesting pattern of clad distortion, seen for 570°F samples (Fig. 8) is not likely to be seen in samples whose clad has a larger radius of curvature. The "caterpillar" appearance shows, however, how clad distortion proceeds through cycles of bulging and splitting.

C. Progress of Failure in Defected Sections from Full-Diameter Tubes

Sections from full diameter tubes have been used to obtain quantitative data on the progress of corrosion from an intentionally inserted cylindrical defect. Such cylindrical holes are readily inserted in the cladding and have been used in various laboratories for such studies,[9-12] although there may be some question regarding how realistically they resemble damage likely to occur during reactor service. In this study, hydrogen evolution is taken as the measure of corrosion.

Sections about 6-inches long are bored out at the ends and end seals are welded in. After a test to check weld integrity, the defect is inserted. In all the tests discussed here, the defect has remained set as a cylindrical hole whose diameter and depth are both 40 mils. After the autoclave containing the assembly has been brought to the temperature of the test, gas is bled periodically from the autoclave. The steam is condensed and the hydrogen passes through a wet test meter, which thus provides a record at any time of total hydrogen evolved and, therefore, of total uranium converted to oxide. (The consistency of hydrogen evolution data with weight loss data was shown in a series of experiments with bare end specimens. A conveniently applied conversion factor of 5 grams U/liter H_2 measured at room temperature is considered sufficiently accurate.) Plots of these data as a function of time then represent the progress of corrosion. Figures 9, 10 and 11 show such plots for runs at 530°F (277°C), 570°F (299°C) and 660°F (349°C) respectively. Samples were run either as-extruded or following the standard diffusion anneal (7 hours at 880°C).

These plots can be used to deduce the extent of corrosion during specified intervals at the same temperature following achievement of a corrosion level which would be used as the basis for detection of failure in a reactor. The signal designated as necessary for detection of failure may be based on a specified total weight of uranium converted to UO_2. Thus, the plots are used to establish the time $T_{s(2.5)}$ or $T_{s(5)}$ at which 2.5 or 5 grams has been oxidized. The plots may then be read to determine how much more uranium is converted to oxide during various intervals after the respective Ts. Data obtained in this way are given in Tables IV and V for 530°F and 570°F respectively. The 660°F data of Figure 11 do not lend themselves to such evaluation of the signal time and of corrosion behavior after receipt of a signal. Attack was apparently well underway by the time the test temperature was reached.

Evaluation of the data in Tables IV and V show that at these temperatures the diffusion heat treatment provides an added margin of safety. For various intervals after the detection of failure, heat treated material undergoes less attack. These results remove one of the possible objections to the diffusion anneal. Fear has been expressed that although the anneal might indeed reduce the frequency of failure, it might also constrain failure initiated at a defect too large to be overcome by the heat treatment. Corrosion might then proceed slowly and undetected to the point where the constraint would be overcome abruptly, leading to rapid, catastrophic failure. The data presented here show that the suggested pattern of constraint and rapid failure is not found with the cylindrical defect at these temperatures. Instead, heat treatment reduces the damage to fuel elements which remain in the reactor for various intervals after receipt of a signal. The first runs in 660°F (349°C) water, depicted in Fig. 11, indicate that the advantage of heat treatment has been lost at this temperature and that heat treatment may even accelerate failure somewhat.

At 570°F, heat treated tubes show evidence of an intermittent type of local constraint. The pattern of hydrogen evolution is at times erratic. Abrupt rises in rate are seen, but are followed by a leveling off rather than an acceleration. Some uranium is believed to have undergone conversion to hydride by reaction either with the water or with hydrogen evolved in corrosion of other uranium. Uranium undergoing such conversion to hydride obviously is not releasing hydrogen to the wet test meter. After a while, however, a local burst of the cladding may expose this hydrided uranium to water, giving a rapid release of hydrogen. The generation of hydrogen then subsides. Further work is necessary on the role of hydrogen in the progressive failure. The present method of periodically bleeding evolved hydrogen prevents hydrogen from building up. Work on the role of hydrogen is planned with both defected specimens and with transverse section having exposed ends.

The appearance of samples corroded at the three test temperatures is shown in Fig. 12, 13 and 14. The marked change in the nature of attack as a result of heat treatment is perhaps even more striking than for small rod specimens (Figs 4 and 5). In as-extruded material, the main progress of corrosion is longitudinal(parallel to the axis). The build-up of corrosion product causes bulging of the cladding, which ruptures and permits repetition of the cycle of corrosion, bulging and rupture of the cladding farther away. The over-all effect then is damage to the cladding over a considerable area. In heat treated material, the bulging is confined, even at 660°F, to a smaller area around the defect. Corrosion penetrates into the core and is more likely to reach the inner cladding and even to cause this cladding to bulge inward and rupture. Such bulging is not expected to cause significant blocking of flow of coolant through the tube, but as-extruded material is likely to become more difficult to remove the process tube.

D. <u>Failure of Exposed Transverse Sections of Tubes</u>

Tests of transverse sections with the core exposed only at the flat ends have been discussed previously for determination of corrosion rates of core alloys. Such sections may be helpful in the study of the mechanism of corrosion of clad tubes and of the factors affecting such corrosion, although tests involving such surfaces are not likely to represent any failure occurring during reactor operation.

The point of departure for such tests is a 3 hour test in 570°F water (299°C) of an end of heat treated Tube 9-1. Hydrogen was bled periodically as in the defect tests. At the end of this test, the cladding was found intact. It looked as if the core had been etched out to a depth of 1/8 inch, corresponding to an observed weight loss of about 2 $g/cm^2/hr$ (see Table III).

Further work is necessary to establish whether such preservation of clad integrity can be expected consistently under these conditions. The role of hydrogen has to be considered. Damage to the cladding may occur within 3 hours at this temperature if excess hydrogen is present or even if the generated hydrogen is not bled off.

V. SUMMARY AND CONCLUSIONS

A standard diffusion heat treatment for 7 hours at 880°C was established to overcome the effect of subtle defects in 15-mil Zircaloy-2 cladding coextruded over a core of U-2 $^{w}/o$ Zr. This heat treatment has been found to change the nature of failure resulting from gross defects. Corrosion of heat treated material tends to be more confined to the area around and under the defect. Quantitative data on the progress of corrosion have been obtained by continual measurement of the hydrogen evolved. These data on samples with cylindrical defects tested in water at 530°F (277°C) or 570°F (299°C) have shown that heat treatment leads to slower corrosion in various periods after corrosion has reached the level necessary for detection. In the first runs in 660°F (349°C) water, heat treatment led to more rapid failure.

The heat treatment was not found to produce and adverse effects on other properties of the Zircaloy-2 cladding. The effectiveness of the heat treatment can be impaired if the material is cooled too slowly after heat treatment. Temperatures in the neighborhood of 600°C have to be avoided.

Further work is necessary to establish the pattern of corrosion resulting from defects other than the cylindrical holes which have been emphasized thus far. Samples have to be tested under a set of conditions simulating those expected in a reactor shutdown following receipt of a signal. Work on the mechanism of corrosion will emphasize the role of hydrogen.

VI. REFERENCES

1. A. R. Kaufmann, J. L. Klein, P. Loewenstein, and H. F. Sawyer, "Zirconium Cladding of Uranium and Uranium Alloys by Coextrusion", Report NMI-TJ-8 in TID-7546, Book I, Fuel Elements Conference, Paris, November 18-23, 1957.

2. F. H. Lamartin, Jr., and S. Levine, Corrosion Prevention in Clad Nuclear Fuel Rods, S.M. Thesis, Massachusetts Institute of Technology, 1955.

3. F. H. Lamartin, Jr., and S. Levine (R. G. Jenkins), Corrosion Prevention in Clad Nuclear Fuel Rods, in Technical Progress Report for the Period April 1955 through September 1955, USAEC Report NMI-1139, Nuclear Metals, Inc., 1956.

4. R. G. Jenkins, NMI-1152, "Improved Corrosion Resistance of Zircaloy-2 Clad Rods Subjected to Diffusion Anneal", in Metallurgy Information Meeting, TID-7526 (Part 3), p. 259, Ames Laboratory, Iowa State College, May 1956.

5. E. Epremian, Ed., Nuclear Fuels Newsletter, USAEC Report WASH-703, USAEC Division of Research, 1957, p. 30.

6. E. Epremian, Ed., Uranium Alloy Newsletter, USAEC Report WASH-701, USAEC Division of Research, 1957, p. 8.

7. Y. Adda, et al, Étude de la Diffusion Uranium-Zirconium, Report CEA-672, Commissariat à l'Énergie Atomique, France, 1957.

8. A. R. Kaufmann, Visit to Saclay, Nuclear Metals, Inc. internal memorandum, December 4, 1957.

9. R. F. S. Robertson and F. H. Krenz, Defect Test on U - 2 w/o Zr Alloy in the X-2 Loop, Test No. 1, Report CRDC-646, Atomic Energy of Canada, Ltd., 1956.

10. R. F. S. Robertson, Defect Test on U - 2 w/o Zr Alloy in the X-2 Loop, Test No. 2, Report CRDC-647, Atomic Energy of Canada, Ltd., 1957.

11. K. D. Hayden, Summary Report of Fuel Element Rupture Tests in High Temperature, High Pressure Recirculating Water Systems, USAEC Report HW-48836, Hanford Atomic Products Operation, 1957.

12. C. L. Angerman and E. C. Hoxie, Pinhole Corrosion Testing of Aluminum-Clad, Nickel-Bonded Fuel Elements, USAEC Report DP-269, E. I. du Pont de Nemours and Co., 1958.

VII. APPENDIX A - Effect of Diffusion Heat Treatment on Concentration of Uranium in Zircaloy Cladding

One of the undesirable effects which may result from the diffusion heat treatment is an excessive increase in the concentration of uranium at and near the surface of the Zircaloy cladding.

The uranium concentration in the cladding was determined by analysis of successive layers of Zircaloy chips machined (or, in some cases, pickled) from different rod samples, as extruded and after heat treatments longer than those necessary for protection against subtle defects. A fluorometric method was used for analysis.[1] To increase the sensitivity of the method to about 1 ppm, the sample size was increased to the 0.5-gram range by increasing either the length of the sample or the thickness of the analyzed layer. (It should be noted that for rods with a diameter in the 0.3-0.4 inch range, a one-mil layer from a one-inch length weighs about 0.1 gram.) Thus, in addition to individual one-mil layers, samples combining the 4 outermost mils were taken.

The data in Table VI were obtained in this way for samples from two different rods. The analytical method is a relative one, giving the increase in uranium concentration above a reference material such as some Zircaloy stock (in this case, SMZ-556). Absolute determination of the uranium content in other Zircaloy stock (SMZ-387) was attempted with the diethyl ether extraction method used by Westinghouse (WAPD-CTA(GLA)-431). This determination indicated that SMZ-387 extruded stock contained 3-5 ppm U. Analysis of SMZ-387 relative to SMZ-556 showed the latter to contain about 2 ppm U less than SMZ-387, leading to the inference that SMZ-556 contained 1-3 ppm U. Since the rods T and U, listed in Table VI, were prepared from SMZ-387, it is apparent that the surface of these rods had essentially the same uranium concentration as the SMZ-387 stock used in their preparation. The data of Table VI provide no evidence that heat treatment increases the uranium concentration in the outermost four mils of Zircaloy.

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1. Frederick A. Centanni, Arthur M. Ross, and Michael A. DeSesa, "Fluorimetric Determination of Uranium" in Analytical Chemistry, Vol. 28, p. 1651, November, 1956.

Attempts to calculate the surface concentration of uranium were made by Kenneth B. Larson at NMI. He used diffusion coefficients reported elsewhere and also extrapolated analytical data obtained here for Zircaloy layers closer to the core. A difference by a factor of about six between two diffusion coefficients reported for uranium in zirconium led to widely different values for the surface concentration. Extrapolation of analytical data showed that the increase in surface uranium concentration resulting from an 8-hour heat treatment at 900°C was in the range of several tenths of a part per million.

VIII. APPENDIX B - Effect of Diffusion Heat Treatment on Corrosion Resistance of Zircaloy Cladding

Two sets of experiments demonstrated the absence of any adverse effect of diffusion heat treatment on the Zircaloy cladding's corrosion resistance. The earlier set gave qualitative results on short rod samples with welded end plugs, similar to those used for defect tests. Heat-treated samples had a satisfactory appearance following 750°F steam (1500 psi) tests, but the heavy corrosion around the welds prevented evaluation on the basis of relative weight gains. To obtain a quantitative evaluation of cladding corrosion rates resulting from heat treatment, a set of clad rods about 8 inches long was coextruded with integral end seals, eliminating the need for welding prior to corrosion tests. Two such rods were obtained per extrusion. The weight gains (Table VII) and the black corrosion film for both the clad rods and sections of Zircaloy cropped between the uranium alloy core portions show the absence of any adverse effect due to heat treatment.

These results are consistent with a recent report of a systematic study of the effect of heat treatment on the corrosion resistance of Zircaloy-2.[1] Heating temperature was varied in steps of 100°F from 1400 to 1900°F. The best temperatures were found to be 1400 and 1600°F. It was also found that the best cooling treatment is air cooling; the worst is furnace cooling. Goodwin's conclusions indicate that the diffusion treatment at 880°C (1616°F) will not adversely affect the Zircaloy-2 as such (leaving uranium diffusion out of consideration).

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1. J. G. Goodwin, "The Effect of Heat Treatment on the Corrosion Resistance of Zircaloy-2 and Zircaloy-3", Report WAPD-BT-6, Bettis Technical Review, January, 1958.

IX. APPENDIX C - Effect of Diffusion Heat Treatment on Microstructure of Zircaloy

Heat treatment of Zircaloy at temperatures high in the alpha region or low in the alpha-beta region causes coarsening of the Zircaloy grains. In addition, slow cooling causes excessive precipitation of zirconium intermetallics with the alloying elements. Such precipitation can adversely affect the Zircaloy's corrosion behavior.

Figures 15 and 16 show photomicrographs of sections from tube 9-1 and tube 12. (The different magnifications in the two figures should be noted.) Tube 9-1 was part of a tandem extrusion and was heat treated vertically. Tube 12 was a full-length tube and was heat treated horizontally. The difference in grain size for the Zircaloy in the two tubes bears no obvious relation to the method of heat treatment. This different may be due to a difference in composition, such as in hydrogen content. Even in tube 12, however, coarse grains are seen near the core-cladding interface but not near the surface. Coarse grains do not extend across the full width of the cladding.

Intergranular precipitation of intermetallics is not heavy and apparently has not impaired corrosion behavior (Appendix B). The samples shown here resemble one shown by Goodwin[1] (his Figure 9) for a 1600°F (871°C) heat treatment followed by air cooling.

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1. J. G. Goodwin, "The Effect of Heat Treatment on the Corrosion Resistance of Zircaloy-2 and Zircaloy-3", Report WAPD-BT-6, Bettis Technical Review, January, 1958.

X. TABLES AND FIGURES

Table I

Effect of Different Heat Treatments On Intentionally Defected Specimens

Corrosion Test in 650°F (343°C) Water

Each sample contains four 7-mil diameter defects drilled through Zircaloy-2 cladding to U - 2 w/o Zr core; all samples air-cooled after heat treatment.

Heat Treatment Temp., °C	Duration of Heat Treatment, Hr	Samples Failed		Number of samples which completed more than 1000 hr in test before withdrawal
		Number	Interval of Failure, Hr	
825	9	1 1* 1	4 - 44 8 - 77 726 - 936	1*
	12	1 1	66 - 279 936 - 1099	2*
	16	1	84 - 102	3
	25			4
880	5	1	4 - 265	4
	6	1	0 - 4	9
	7			10**
	8			7
	9			4
900	1	4	0 - 4	
	2	1	0 - 4	3
	4	1	0 - 67	10
	5			19
	6			6

*Only one defect in specimen
**Also see data of Table II

Table II

Effect of Different Thermal Histories After the 7-Hour 880°C Diffusion Treatment

Corrosion Test in 650°F (343°C) Water

Each sample contains four 7-mil diameter defects drilled through Zircaloy-2 cladding to U - 2 w/o Zr core.

	Samples which Failed	Interval of Failure, Hr	Samples which completed more than 1000 hr in test before withdrawal
Air cooled from 880°C to room temperature			T-27; U-27*
Furnace cooled from 880°C to 650°C; air cooled			T-22; T-24
Furnace cooled from 880°C to 600°C; air cooled	T-26; U-26	4 - 78	
Furnace cooled from 880°C to 550°C; air cooled	T-23; T-25	0 - 4	
Air cooled; 1/4 hr at 600°C; air cooled			T-30; U-30
Air cooled; 1/2 hr at 600°C; air cooled			T-32; U-32
Air cooled; 1 hr at 600°C; air cooled	T-31	4 - 78	U-31
Transferred to 690°C furnace; 1 hr at 690°C; air cooled			T-7; U-7
Transferred to 690°C furnace; 2 hr at 690°C; air cooled	T-10	4 - 121	U-10
Transferred to 690°C furnace; 4 hr at 690°C; air cooled	T-8; U-8	4 - 121	
Transferred to 690°C furnace: 6 hr at 690°C; air cooled	T-9 U-9	4 - 121 0 - 4	
Air cooled; furnace cooled from 550°C			T-33; U-33

*These samples were not included in Table I.

Table III

Corrosion Rates of Uranium-Zirconium Alloys
$gm/cm^2/hr$

Temp.		Unalloyed U			U-2 $^w/o$ Zr		U-5 $^w/o$ Zr		U-7.5 $^w/o$ Zr	U-10 $^w/o$ Zr		U-15 $^w/o$ Zr
^oC	^oF	NMI	BMI-998	CRDC-646 (from CT-3047 and CT-3055)	NMI	CRDC-646	NMI	BMI-1156	NMI	NMI	BMI-1156	NMI
100	212	0.003	0.003	0.003	0.001	0.003 0.0017 (Ames- (WASH- 153)	0.001	0.0013 (Ames)	0.0007	0.0003	0.000051 (Ames) 0.001	
150	302			0.036		0.036						
200	392			0.23		0.23		0.16			0.06	
227	440	2.0	0.9	0.7	0.6	0.4	0.6		0.5	0.36		
250	482			2		1.0						
260	500	3.0	1.4		1.0	1.1	1.0	1.0	1.0	0.7	0.5	0.5
277	530				1.3	1.5						
299	570				2.0	3.0						
316	600		10.0 5-10 (BMI- 1156)		3.0	5.0		5.0			2.0	
343	650				6.0	7.0	6.0	10.0	2.5	(7)	5.0	

Table IV

Tests of Intentionally Defected Tube Sections
Defect: 40 mils diameter; 40 mils depth

Series	Condition	Material	Temperature
3-10	As Extruded	U - 2 w/o Zr clad with 15 mils of Zircaloy-2	530°F (277°C)
7H2	Heat Treated		

Specimen	Date	$T_{s(2.5)}$* (hr)	Grams Uranium after $T_{s(2.5)}$ + x hr							T_t (hr)**	$T_t - T_s$
			x = 1	x = 2	x = 3	x = 4	x = 6	x = 8	x = 16		
3-10-A	4/8	7.75	0.5	1.5	3.7	15.3				12.25	4.5
3-10-B	4/9	6.30	13.3							7.75	1.5
3-10-C	4/16	10.50	1.0	9.5	22.0	38.0				15.25	4.8
3-10-D	4/22	4.80	13.8	52.8	118.8					8.00	3.2
7-H2-A	4/11	6.00	2.0	3.5	6.0	8.5	15.0	23.5		14.00	8.0
7-H2-B	4/18	6.00	1.0	2.0	3.5	5.0	13.0	19.0		19.75	13.8
7-H2-C	4/21	5.30	1.5	3.7	6.0	8.8	14.8	22.0	62.5	25.50	20.2
		$T_{s(5.0)}$ (hr)	**Grams Uranium after $T_{s(5.0)}$ + x hr**								
3-10-A	4/8	10.50	8.8							12.25	1.8
3-10-B	4/9	6.65	21.1							7.75	1.1
3-10-C	4/16	11.75	9.5	23.5	39.1					15.25	3.5
3-10-D	4/22	5.10	21.1	70.0	150.0					8.00	2.9
7-H2-A	4/11	7.40	2.0	4.0	7.0	10.3	18.5			14.00	6.6
7-H2-B	4/18	8.40	1.8	3.0	7.5	12.0	17.5	23.8		19.75	11.4
7-H2-C	4/21	6.75	2.3	4.8	7.5	10.5	17.5	25.0	66.3	25.50	18.8

*$T_{s(xx)}$ = time at which a signal is received corresponding to (xx) grams of total uranium oxidized

**T_t = termination time of test.

Table V

Tests of Intentionally Defected Tube Sections
Defect: 40 mils diameter; 40 mils depth

Series	Condition	Material	Temperature
4	As Extruded	U - 2 w/o Zr clad with 15 mils of Zircaloy-2	570°F (299°C)
7H2	Heat Treated		

Speci-men	Date	$T_{s(2.5)}$* (hr)	Grams Uranium after $T_{s(2.5)}$ + x hr						T_t (hr)**	$T_t - T_s$
			x = 1	x = 2	x = 3	x = 4	x = 6	x = 8		
4B	5/2	3.3	7.5	30.0	75.0	155.0			7.25	4
4C	5/5	1.6	12.5	52.5					4.40	2.8
4D	4/23	2.7	7.0	55.0	127.5				5.75	3
7H2D	4/29	3.1	3.0	6.0	37.0	45.3	65.0	85.3	12.75	9.6
7H2E	4/23	2.5	2.5	5.5	9.5	15.75	36.5	61.0	11.75	9.2
7H2F	5/2	2.3	2.25	5.5	20.0	27.0	41.0		9.67	7.4
		$T_{s(5.0)}$ (hr)	Grams Uranium after $T_{s(5.0)}$ + x hr							
4B	5/2	3.75	13.0	46.0	100.0				7.25	3.5
4C	5/5	1.9	17.5	67.0					4.40	2.5
4D	4/23	3.2	17.5	87.5					5.75	2.5
7H2D	4/29	4.0	3.25	32.5	41.5	51.0	71.3	93.4	12.75	8.8
7H2E	4/23	3.5	3.0	7.0	13.25	19.0	43.5	106.5	11.75	8.2
7H2F	5/2	3.4	3.25	18.0	25.0	32.0	76.5		9.67	6.3

*$T_{s(xx)}$ = time at which a signal is received corresponding to (xx) grams of total uranium oxidized

**T_t = termination time of test.

Table VI

Increase in Uranium Concentration (ppm) in Successive Layers of Zircaloy-2 from As-Extruded and Heat-Treated Rod Samples Reference Material SMZ-556

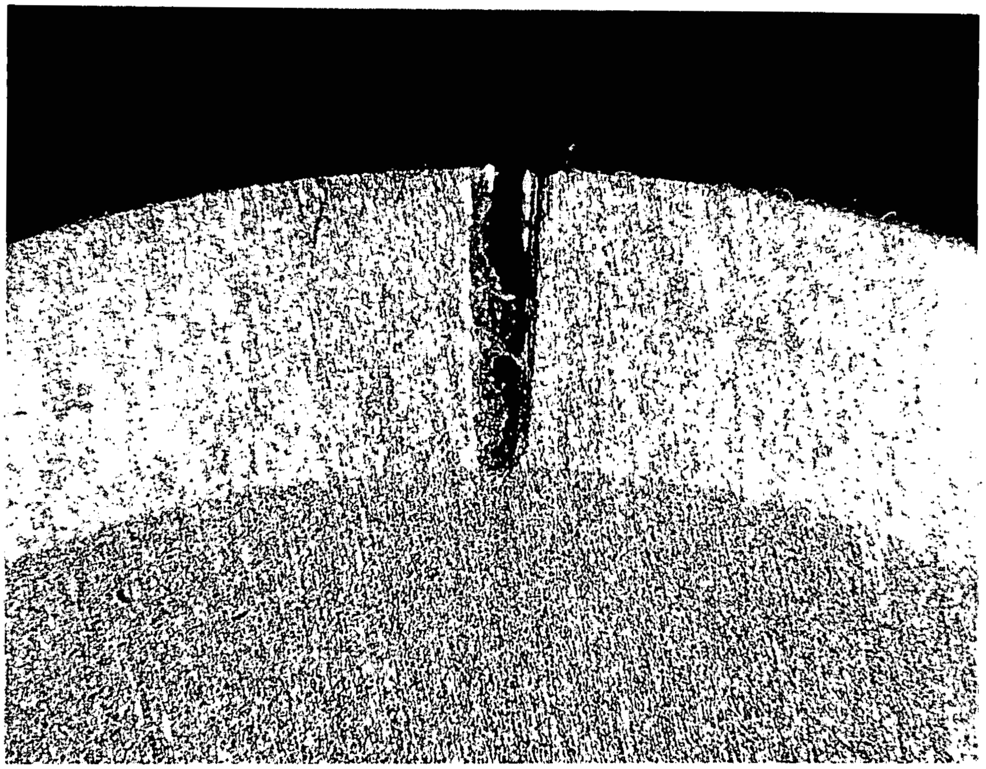
Depth of Zircaloy	T			U		
	As Extruded	9 hr/880°C	8 hr/900°C	As Extruded	9 hr/880°C	8 hr/900°C
Outermost mil	8.87	20.13	12.32	889.0	1.88	1.19
Second	0.92	5.81	6.49	0.30	1.92	2.11
Third	6.79	2.82	1.76	0.0	0.33	1.46
Fourth	0.33	1.37	2.15	0.08	0.26	0.91
Fifth	3.43	1.15	0.44	0.73	1.32	3.15
Combined sample from outermost four mils	1.99	0.98	18.84	0.80	1.04	1.39

Table VII

Corrosion Tests in 750°F Steam of Zircaloy-2 Cladding From Clad Rods With Integral End Seals

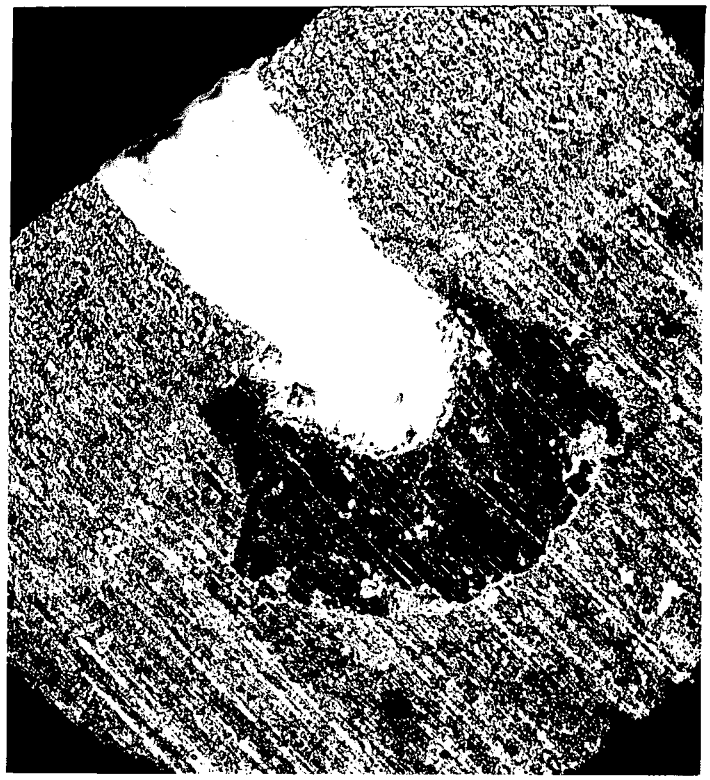
Zircaloy-2 Stock SMZ-387

	Weight Gains, mg/dm^2, in 750°F Steam			
	3 Days		14 Days	
	Clad Rods	Zircaloy Cropped Between Cores	Clad Rods	Zircaloy Cropped Between Cores
As Extruded	18;18	18;20	31;31	27;28
9 hr/880°C, air cooled	13;14	24	27;24	35
8 hr/900°C, air cooled	18;22	23	32;35	31



50X RF-4579

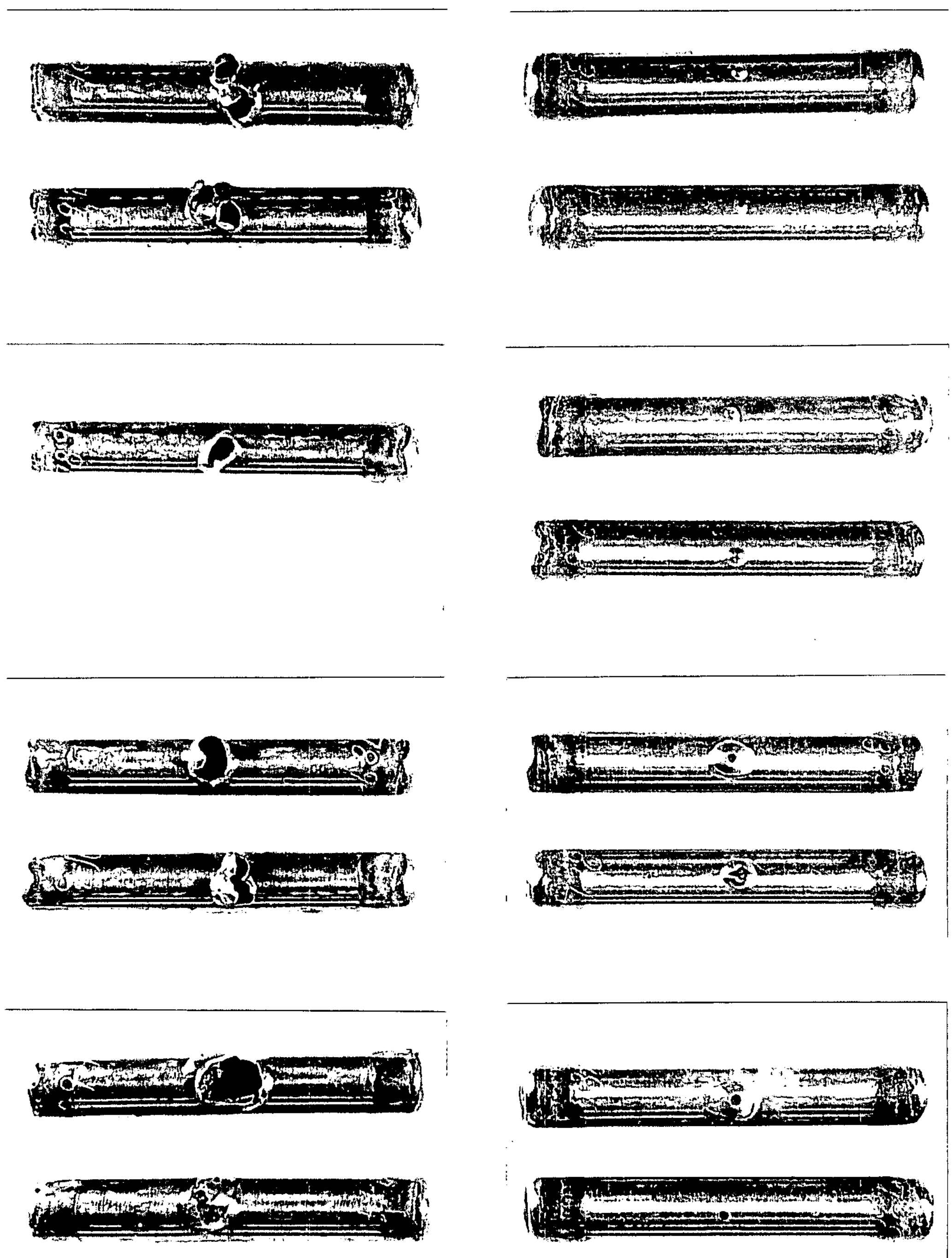
Fig. 1 - "Automatic" Drilling. 7-mil defect through 30-mil Zircaloy. (Defect drilled in as-extruded rod.)



100X A-2045-1

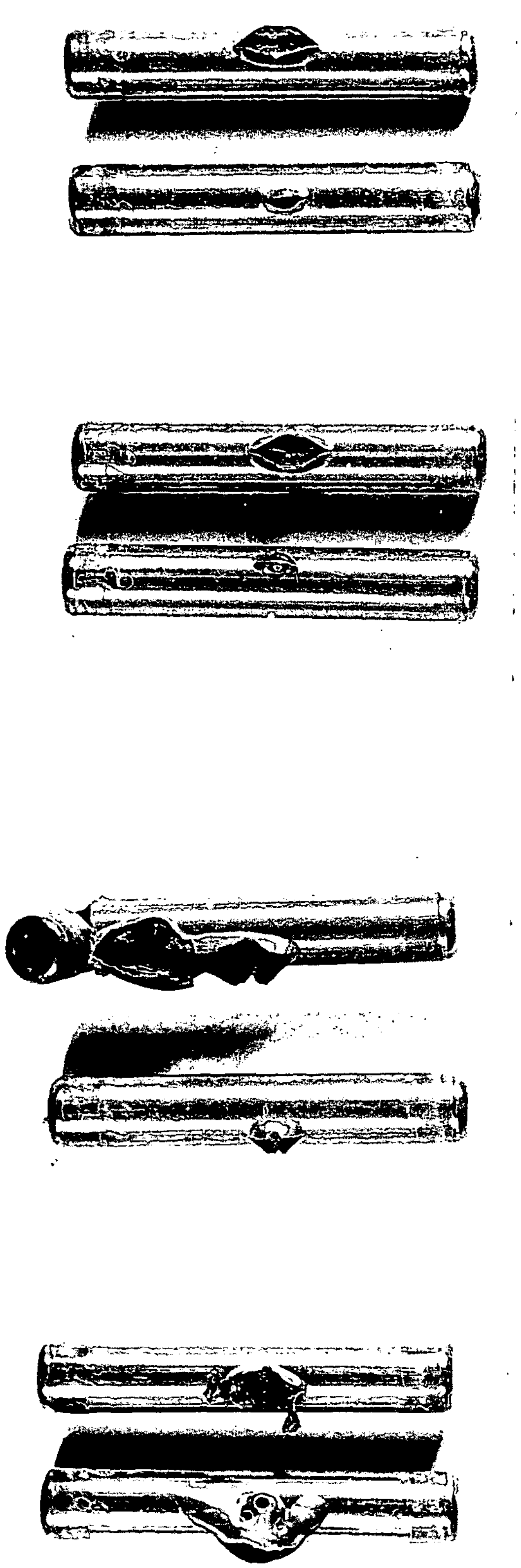
Fig. 2 - Section through Defect No. 2 of Tube 9-1.
Defect: diameter, 7 mils; original depth, 16 mils.
Tested 32 hours in 600°F (316°C) water.

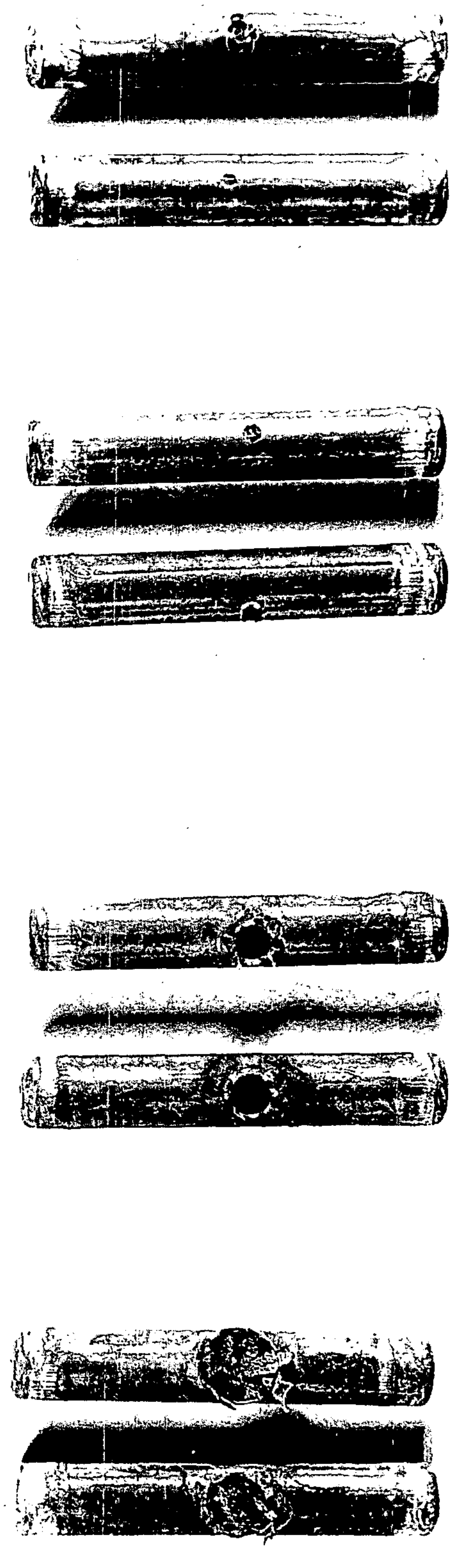
Note accumulation of corrosion product (black) around base of defect.



RF-5392

Set A   Set B





RF-5397

Set C

Set D

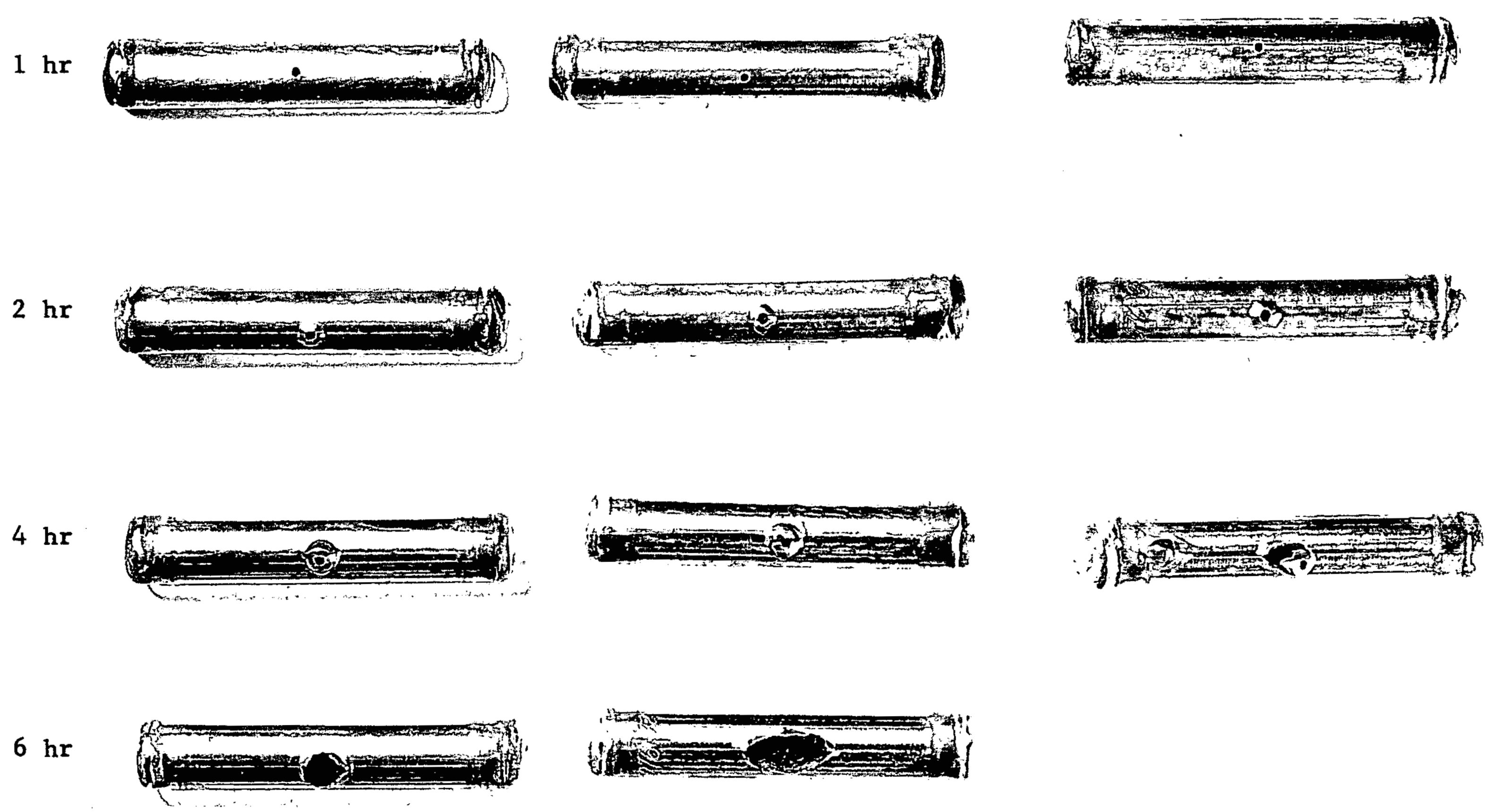
As Extruded

Heat Treated - 10 min/800°C

Heat Treated - 15 min/880°C

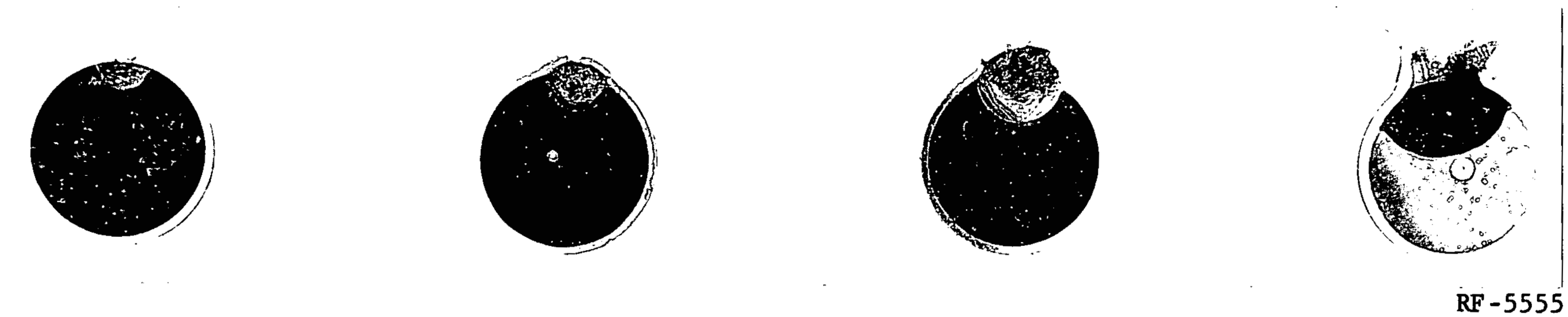
Heat Treated - 7 hr/880°C

Fig. 5 - Effect of Various Heat Treatments on Corrosion. Defect Diameter - 40 mils. Depth - 40 mils. Corrosion Tested - 4 hr/530°F. Nominal length of each sample, 2 inches. RF-5701.



RF-5546

Fig. 6 - Progress of Corrosion at 530°F (277°C). Defect Diameter - 40 mils. Depth - 40 mils. Nominal length of each sample, 2 inches.



1 hr   2 hr   4 hr   6 hr

<u>Fig. 7</u> - Section Through Defect of Samples Corroded at 530°F (277°C).
Defect Diameter - 40 mils. Depth - 40 mils. See Fig. 6.
Magnification approximately 3X.

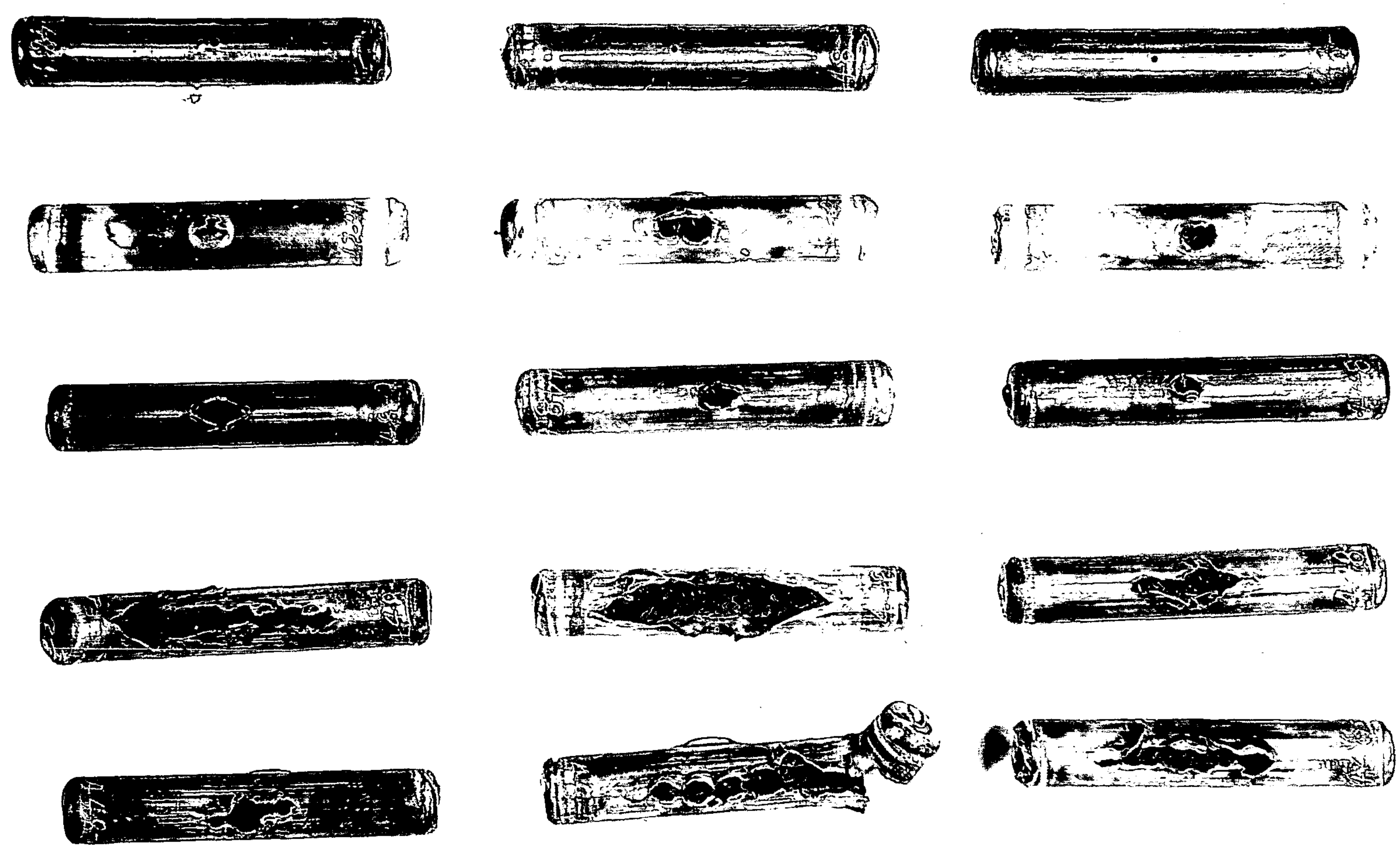
0 hr

1/2 hr

1 hr

1-1/2 hr

2 hr



RF-5576

Fig. 8 - Progress of Corrosion at 570 °F (299 °C). Defect Diameter - 40 mils. Depth - 40 mils. Nominal length of each sample, 2 inches.

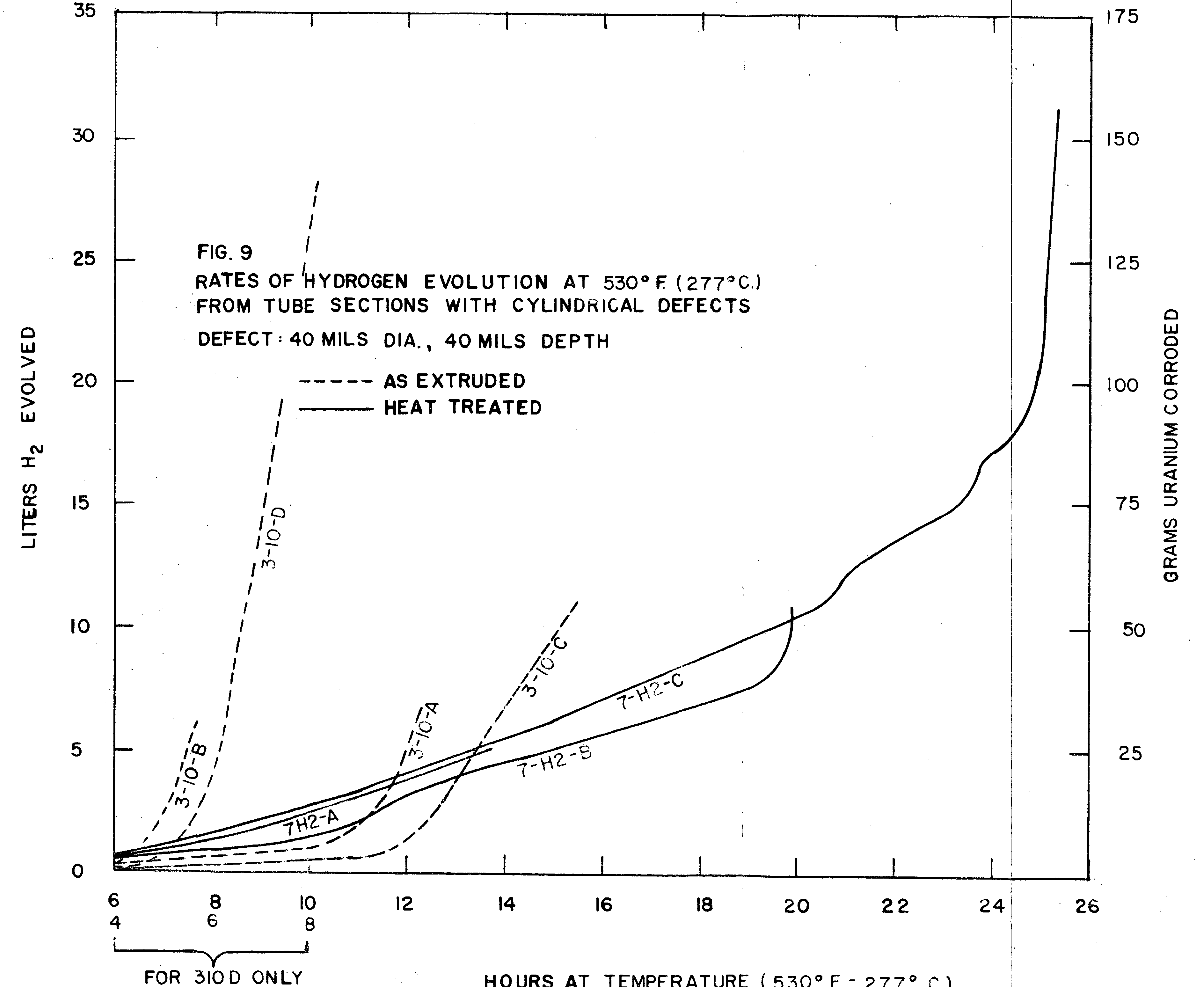


FIG. 9
RATES OF HYDROGEN EVOLUTION AT 530° F. (277°C.)
FROM TUBE SECTIONS WITH CYLINDRICAL DEFECTS
DEFECT: 40 MILS DIA., 40 MILS DEPTH
AS EXTRUDED
HEAT TREATED
LITERS H_2 EVOLVED
GRAMS URANIUM CORRODED
HOURS AT TEMPERATURE (530° F - 277° C.)
FOR 310 D ONLY
3-10-D
3-10-B
3-10-A
3-10-C
7H2-A
7-H2-B
7-H2-C
0
5
10
15
20
25
30
35
25
50
75
100
125
150
175
6
8
10
12
14
16
18
20
22
24
26
4
6
8

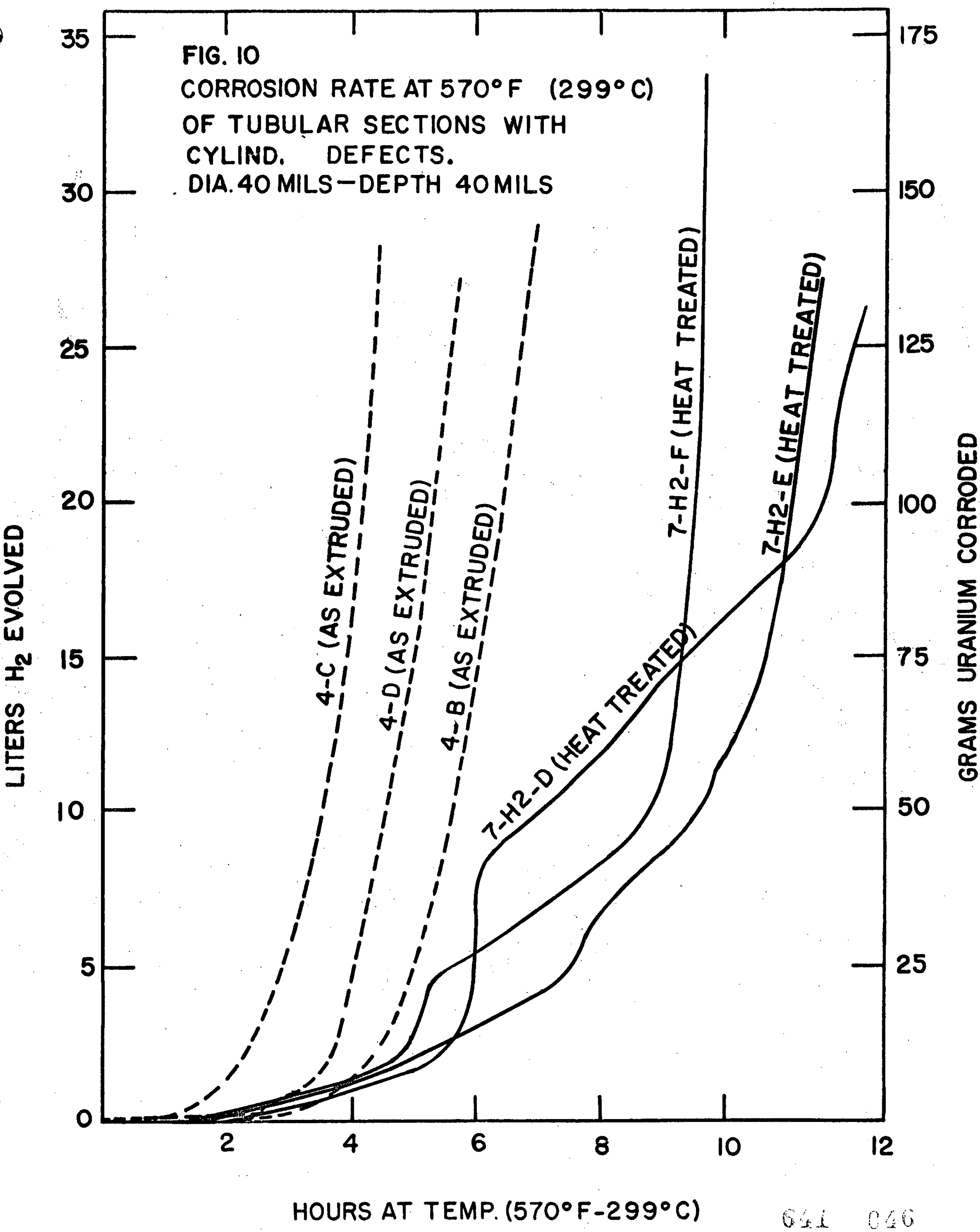


FIG. 10
CORROSION RATE AT 570° F (299° C)
OF TUBULAR SECTIONS WITH
CYLIND. DEFECTS.
DIA. 40 MILS—DEPTH 40 MILS
4-C (AS EXTRUDED)
4-D (AS EXTRUDED)
4-B (AS EXTRUDED)
7-H2-D (HEAT TREATED)
7-H2-F (HEAT TREATED)
7-H2-E (HEAT TREATED)
LITERS H2 EVOLVED
GRAMS URANIUM CORRODED
HOURS AT TEMP. (570° F-299° C)
0
5
10
15
20
25
30
35
25
50
75
100
125
150
175
2
4
6
8
10
12

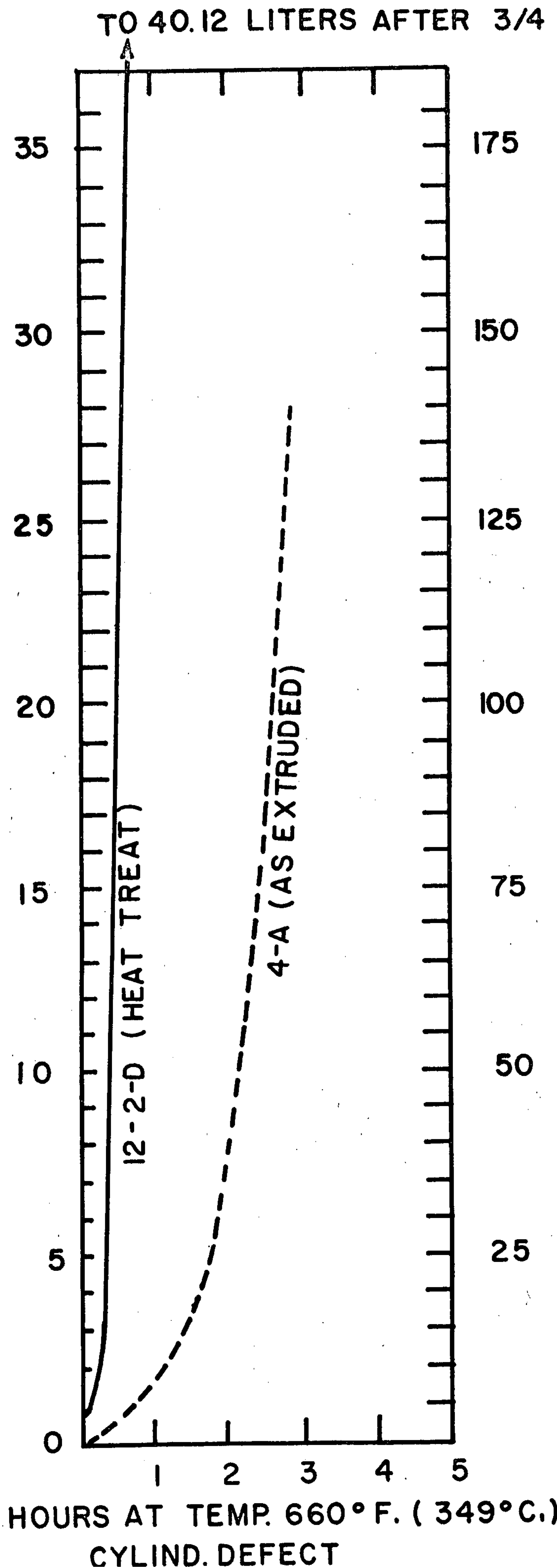


FIG. 11
CORROSION RATE AT 660° F. (349°C.) OF TUBULAR SECTIONS WITH CYLIND. DEFECTS. DIA. 40 MILS, DEPTH 40 MILS

641 047

RF-5651

As Extruded
3-10-A
12-1/4 hr at 530^{o}F
4.5 hr after $T_{s(2.5)}$=7-3/4 hr
1.8 hr after $T_{s(5)}$=10-1/2 hr

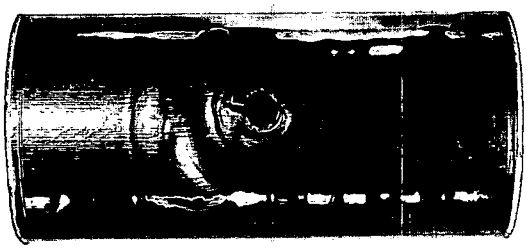
RF-5653

Heat Treated
7 hr/880^{o}C - Air Cooled
7-H2-B
19-3/4 hr at 530^{o}F
13.8 hr after $T_{s(2.5)}$=6.0 hr
11.4 hr after $T_{s(5)}$=8.4 hr

Fig. 12 - Tube Sections Tested in Water at 530^{o}F (277^{o}C). See Table IV and Fig. 9 for corrosion data. Original diameter is 2 inches.

RF-5731

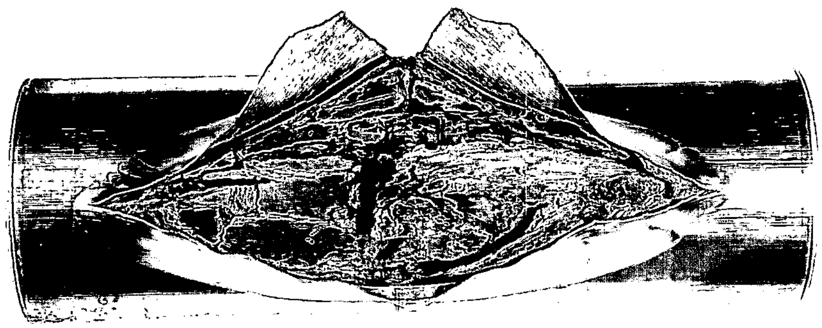
As Extruded
4-B
7-1/4 hr at 570°F
4 hr after $T_{s(2.5)}$=3.3 hr
3.5 hr after $T_{s(5)}$=3.75 hr



RF-5732

Heat Treated
7 hr/880°C - Air Cooled
7-H2-D
12-3/4 hr at 570°F
9.6 hr after $T_{s(2.5)}$=3.1 hr
8.8 hr after $T_{s(5)}$=4.0 hr

Fig. 13 - Tube Sections Tested in Water at 570°F (299°C) Defect diameter - 40 mils. Depth - 40 mils. See Table V and Fig. 10 for corrosion data. Original diameter is 2 inches.



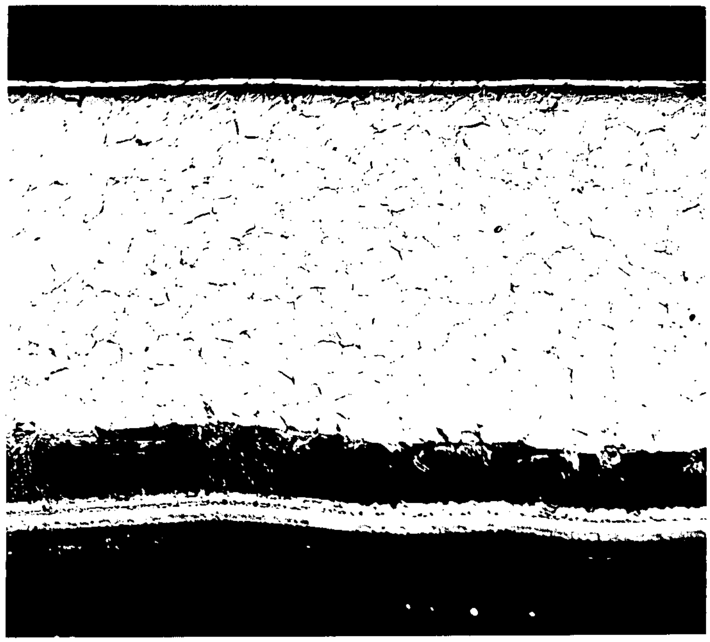
RF-5737

As Extruded
4-A
3 hr at 660°F

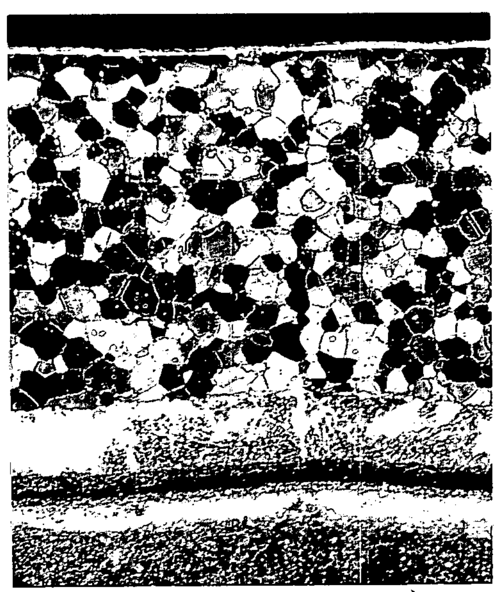
RF-5738

Heat Treated
7 hr/880°C - Air Cooled
12-2-D
3/4 hr at 660°F

Fig. 14 - Tube Sections Tested in Water at 660°F (349°C). See Fig. 11 for corrosion data. Original diameter is 2 inches.

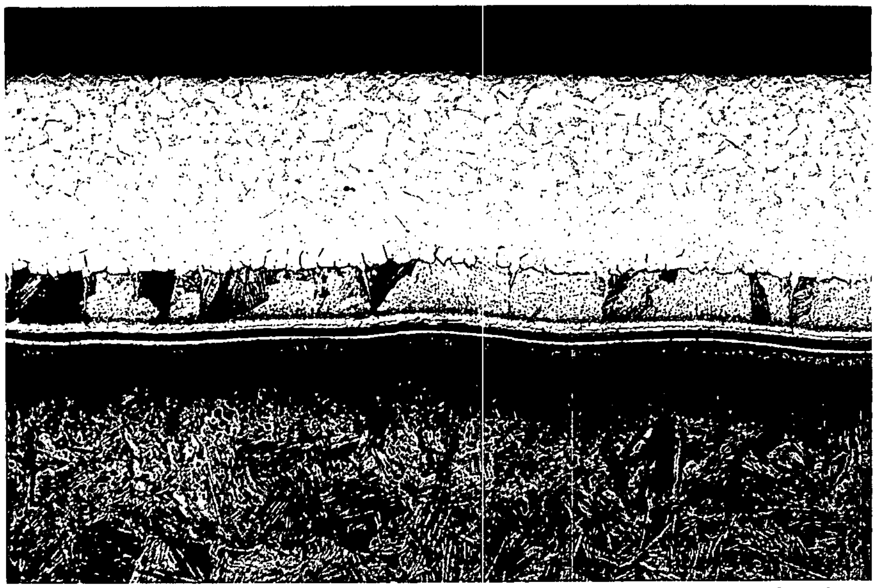


150X Bt. Lt. A-2041-2a

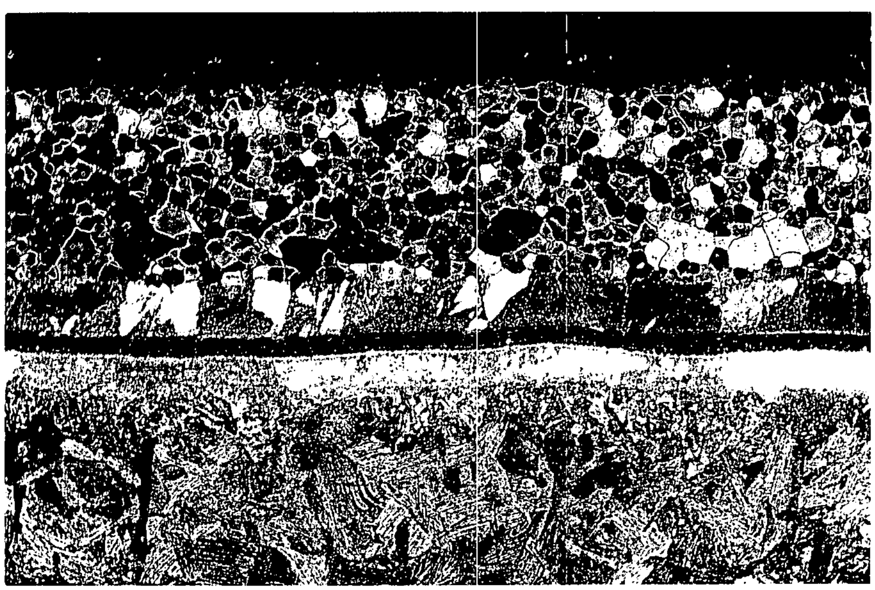


150X Pd. Lt. A-2041-2b

Fig. 15 - Microstructure of Zircaloy-2 Cladding of Tube 9-1 After Standard Diffusion Heat Treatment.



100X Bt. Lt. A-2152-a



100X Pd. Lt. A-2152-b

Fig. 16 - Microstructure of Zircaloy-2 Cladding of Tube 12 After Standard Diffusion Heat Treatment.