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ABSTRACT

Corrosion behavior is a prime consideration in the design of a fuel element to be used in a water-cooled nuclear reactor. Despite their inherently poor corrosion resistance, economically attractive natural uranium or its high alloys can be made into satisfactory fuel elements by use of a sound protective cladding. For U - 2 $^{w}/o$ Zr alloys, the protection offered by the cladding has been increased by the use of a diffusion heat treatment. Despite such protective measures, however, allowance must be made for the occurrence of cladding flaws which would permit exposure of the core material and consequent corrosion, leading to destruction of the fuel element.

Various techniques have been used to study the corrosion of fuel elements with artificial cladding defects in water at temperatures up to 349°C (660°F). These studies have indicated that even in 349°C (660°F) water, the corrosion of certain defected fuel elements is slow enough to permit detection in time for a safe reactor shut down. Such fuel element failure is not expected to be disastrous in terms either of uranium release or of physical damage to the fuel element. Reactors can be designed with appropriate detection systems to permit exploitation of the other advantages of high-uranium metallic fuel elements without concern for the consequences of exposure of the core to water.

INTRODUCTION

The use of high-uranium fuels in a water-cooled reactor may be permissible despite their inherently high corrosion rates. Such fuels can be protected from contact with the water by a sheath or cladding of corrosion-resistant material. The use of clad high-uranium alloys as fuel elements in water-cooled reactors is limited by a lack of faith in the reliability of the cladding and an ignorance of the consequences of exposure of the core material to coolant water.

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* Paper presented by J. Paul Pemsler.

Methods for producing reliable cladding by coextrusion have been developed at Nuclear Metals.[1] Many tubular and rod-type elements with integral end seals have been made whose cladding integrity has been demonstrated. A diffusion heat treatment will increase the protection provided by Zircaloy cladding coextruded with at least one binary alloy, U - 2 $^{w}/o$ Zr. Such a treatment has successfully prevented 343°C (650°F) water from destroying specimens from fuel rods and reactor-size fuel tubes with small holes in the cladding. Additional protection against service failure can be obtained by testing every element and eliminating those with defective cladding before they are used. Allowance has to be made for the possibility that such tests may not detect all cladding defects or that cladding defects may develop during service. Defects of sufficient size will permit the rapid destruction of the fuel element unless preventative steps are taken.

Since rapid corrosion of the core material apparently will be characteristic of metallic fuel element failures in water-cooled reactors fueled with high-uranium alloys, it must be anticipated by reactor designers. Reactors can be designed to cope with a fuel failure and the subsequent release of moderate amounts of activity into the coolant. The early detection of failure is desirable so that coolant contamination by activity can be kept to tolerable levels, and also so that the fuel element can be removed easily and without causing additional damage.

Various techniques for detecting failure have been reported,[2,3,4,5] which basically involve monitoring pressure, temperature, or activity of the coolant water. Reactor designers considering such detection methods have to know the extent and rate of corrosion necessary to give a signal. Other design problems are concerned with the extent of the damage to the element at the time a signal is detected, progress of corrosion following receipt of a signal, and means by which corrosion damage can be minimized.

The purpose of the work reported in this paper has been to find methods for minimizing the frequency of failure of Zircaloy-clad fuel elements of U - 2 $^{w}/o$ Zr, to apply these techniques to actual reactor fuel elements, and to determine the extent of failure when it does occur. Full-size reactor tubes (2.100-inch OD, 1.460-inch ID) with 15 mils cladding, and rods (0.3 to 0.5-inch diameter) with 5, 10, 15, or 30-mil thick cladding have provided the material used for these tests. A large portion of this work has been a part of a program to develop fuel elements for a heavy water-cooled reactor.

DEVELOPMENT OF HIGH-URANIUM FUEL ELEMENTS FOR OPTIMUM CORROSION BEHAVIOR

Corrosion Test Techniques

1. Static Tests - Most of the corrosion tests conducted in this work were made in the standard manner. Specimens, together with a predetermined amount of water, were placed in a pressure vessel (autoclave), which was then

raised to the desired temperature. Air was removed by bleeding off steam. Cooling was generally accomplished by leaving the autoclave in the electrical heating jacket after the power was shut off. More rapid cooling was accomplished by removing the autoclave from the jacket and water cooling it.

A wet test meter, an instrument which measures the volume of gas passed through it, has been used to follow the progress of corrosion initiated at gross cladding defects.[6,7] Steam and the hydrogen from the corrosion reaction are periodically released from the autoclave, the steam condensed, and the hydrogen passed into the wet test meter and its volume determined. The measured volume of hydrogen is then converted to its equivalent in weight of corroded uranium. A factor of approximately 5 grams of U/liter of H_2 at room temperature has been experimentally justified.

It is worth noting that more sophisticated techniques have been used by other investigators to determine the corrosion at a given temperature.

Tuxworth and Krenz[8] have reported several refinements of the standard autoclaving procedure. By using a "drilling autoclave", it was possible to expose the core of sheathed fuel elements to attack by water after the desired temperature was reached. When this technique was combined with an autoclave quenching step, corrosion was permitted to occur only at an essentially constant temperature for a controlled length of time. Measurement of activity released to the water by specimens corroded by the latter technique after a brief irradiation appeared to be a sensitive method for measuring relative corrosion rates.

Grieser and Simons[9] have started corrosion at any selected time by admitting water, preheated to the desired temperature, to the preheated test autoclave charged only with the specimen. Corrosion was stopped fairly rapidly by bleeding off the water after corrosion had progressed to the desired stage. They were able to observe and film the progress of corrosion through observation windows in the autoclave.

2. Dynamic Tests - Flowing steam tests were conducted in a system made available for these tests by the staff at Argonne National Laboratory.[10] The system was made so that steam was continually entering the bottom of the autoclave chamber which contained the specimens. A back pressure regulator in the exit line, nearer to the top of the autoclave, prevented appreciable buildup of gas pressure. Therefore, the rate of hydrogen release should correspond closely with the rate of production as a result of the corrosion reaction.[11]

These and other corrosion test techniques in combination with various specimen types and test conditions have provided a large body of data on the aqueous corrosion behavior of uranium and high-uranium alloys, both clad and bare. Many of these data will be referred to throughout the remainder of this paper.

Corrosion of Bare Uranium Alloys in Water; Effect of Zirconium

Many of the early studies of the aqueous corrosion of uranium and its alloys were concerned primarily with the development of high corrosion resistance. Little information was obtained concerning the corrosion rates of alloys which corroded rapidly. A few alloys such as U - 2 $^{w}/o$ Zr[12] and U - 3.8 $^{w}/o$ Si[13] were investigated in some detail. Much of the early work for uranium-zirconium alloys has been previously summarized.[12] More recently, the rates for the aqueous corrosion of alloys of uranium with 0 to 15 $^{w}/o$ Zr have been determined by more refined techniques up to 343°C (650°F).[6,7,9,14,15] Some typical corrosion rates for various temperatures are shown in Table I.

The corrosion rates of uranium-zirconium alloys increase logarithmically with temperature rise as can be shown by a typical Arrhenius plot.[3,8] Corrosion rates determined from weight losses indicate that at any temperature from 100°C (212°F) to about 300°C (572°F) the addition of 2 $^{w}/o$ Zr reduced the corrosion rate to about one-third that of unalloyed uranium. The effect of small additions of zirconium to the corrosion rate of uranium was vividly shown by relative rates determined by the Canadians[8] from activity increases in water used to corrode mildly irradiated specimens. These data suggest that the 2 $^{w}/o$ Zr reduces the corrosion rate by a factor nearer to ten. Both weight loss and activity increase data show that further additions of zirconium have much smaller effects.

The effects of heat treatments on the corrosion rates of the high-uranium alloys have not been adequately studied. The effects of low temperature heat treatments were briefly studied at one stage of the work discussed in this paper. Aging caused by heat treatment at 450°C (842°F) and 600°C (1112°F) has no apparent effect on the corrosion rate of bare U - 2 $^{w}/o$ Zr. A more complete discussion of this topic appears on page 13.

Despite the effect of alloying additions such as a few $^{w}/o$ Zr, high uranium alloys can be expected to corrode rapidly in water at elevated temperatures. If such alloys are to be useful as nuclear fuels, their contact with the coolant must be prevented by protective sheaths or claddings of a more corrosion-resistant material.

Protection by Coextruded Cladding

Coextrusion has been found to clad uranium and uranium alloys with reliable cladding and also to provide integral end seals. Zirconium and Zircaloy have been applied to a number of uranium alloys by coextrusion.[1]

Many studies have demonstrated that the coextrusion process effectively provides a sound, well-bonded, protective sheath for the less corrosion resistant core material. A considerable amount of effort at Nuclear Metals has been devoted to the study of the origin of defects or inadequacies in the cladding; methods by which such defects can be detected; effects of various defects on the corrosion behavior of the fuel element; and means by which they may be overcome.

Humphrey and Brooks,[16] for example, had to drill 46.5-mil diameter holes in cladding components prior to extrusion to form 30-mil thick cladding in order to obtain core attack by hydrogen or steam. Similar attempts by the present authors[17] have shown that, unless the defect is large enough to be readily visible in the cladding component before extrusion, it will be sealed during the extrusion. If, however, defects are plugged with some foreign material, for example, impure zirconium or ZrO_2, they frequently do not seal and a path is left for entry of coolant to the core.

Threads or veins of core material have on rare occasions been observed which pass through the cladding of extrusions made under improper conditions or improperly designed. Such defects will lead to attack of the core by coolant water. Similar defects were intentionally produced when uranium wire was used to plug holes drilled in the cladding component. These wire plugs formed 7-8 mil diameter uranium stringers from the core to the surface of the Zircaloy in the extruded rods. They led to ultimate attack of the core material during corrosion tests in water or steam above 316°C (600°F).

The occurrence of harmful cladding defects is reduced considerably by proper quality control and inspection of the cladding components before the extrusion billet is made. Nevertheless, it is necessary to establish the soundness of the cladding before the element is placed in a reactor.

A number of techniques for testing cladding soundness have been evaluated. Leutz and Matheson,[18] for example, melted the core, which showed up large defects but which apparently had undesirable side effects. Humphrey and Brooks[16] compared the reaction of steam and hydrogen at elevated temperatures. They were able to increase the sensitivity of the hydrogen test by utilizing the magnetic properties of UH_3 at low temperatures. However, they concluded the steam test was adequate. Experience with corrosion tests of hundreds of specimens with cladding containing tiny artificial defects (7-mil diameter holes) had indicated that water at 343°C (650°F) is satisfactory. Many of the elements now being produced are tested in 343°C (650°F) to 360°C (680°F) water and/or 399°C (750°F) - 1500 psi steam.

Interdiffusion Heat Treatment

1. Concept - The purpose of the interdiffusion heat treatment (diffusion anneal) is to overcome the effect of inadequacies in the cladding which might not be detected by routine inspection or testing.

Lamartin and Levine[19] 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. Core 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 (Fig. 1). Using rods and tubes of Zircaloy-clad U - 2 $^{w}/o$ Zr, the basic technique was developed to permit full-scale heat treatments of 120-lb, 11.5-ft long fuel tubes with 2.100-inch OD and 1.460-inch ID.

2. Development - Studies were made of 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, subsequent thermal history, and irradiation.[6,21,22] 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., 1.5 hours at 950°C (1742°F), is allowed for the interdiffusion necessary to provide protection.

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, despite a normally adequate diffusion anneal. Failure has been prevented in specimens with weak bonds by use of a more severe diffusion treatment. It has been impossible to use a more severe heat treatment to overcome a lack of the fabricated bond, as it affects corrosion behavior.

The severity of the diffusion anneal necessary to overcome the effect of a defect increases as the clad thickness decreases. A minimum clad thickness of perhaps 10 mils may be a necessary condition for even the most severe heat treatment to overcome the effect of a defect. However, even with cladding thinner than 10 mils, failure is usually delayed by a diffusion heat treatment. Cladding thicker than 10 mils can lead to prolonged survival after adequate heat treatment even if defects penetrate more deeply into the core. Perhaps the dependency of corrosion behavior on clad thickness and severity of heat treatment can be explained on the basis of clad and bond strengths. Thinner cladding, because it is weaker, is less capable of resisting stress due to expansion of uranium on conversion to oxide; therefore, in order for the diffusion anneal to be successful, this reduction in cladding stiffness must be compensated for by greater strengthening of the core-cladding bond.

The diffusion anneal has been applied to Zircaloy-clad cores of U - 2 $^{w}/o$ Zr, unalloyed uranium, thorium, or thorium with different alloying constituents[23,24] but has been found beneficial only when the core is U - 2 $^{w}/o$ Zr. Perhaps this is because the bond between the U - 2 $^{w}/o$ Zr and the cladding is strengthened by the heat treatment, while the bonds between the other materials are weakened.

The thermal history after the diffusion anneal may undo the beneficial effect of the diffusion anneal. The effect of the diffusion anneal is counteracted by subsequent exposure to temperatures in the range from about 700 to 550°C, either by slow cooling or subsequent heat treatment. The cause of this sensitive range is not known. However, it serves to emphasize the necessity for rapid cooling after a diffusion treatment.

3. Side Effects - A diffusion anneal may cause changes in the composite fuel element in addition to preventing corrosion failure. Studies have been made to determine the effects of various diffusion anneals on uranium content at the cladding surface, structure of the cladding, strength of the cladding, and the corrosion resistance of both core and cladding.

The cladding adjacent to the core can be hardened and strengthened by the uranium, which diffuses into it. Amounts of uranium as low as 22 ^{w}/o, such as may occur 4 mils into the cladding, can harden and stiffen zirconium.[25] Even 10 ^{w}/o U increased the 0.2% offset yield strength of zirconium by a factor of three or four at 316°C (600°F).

Heat treatment of Zircaloy at temperatures high in the alpha region or 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-2 corrosion behavior. These effects have been observed in cladding which received interdiffusion heat treatments below about 870°C.

Experiments have shown that the standard diffusion anneal does not have a deleterious effect on the corrosion resistance of the Zircaloy cladding. The Zircaloy-2 on rods diffusion annealed at 880°C (1616°F) and 900°C (1652°F) corroded at acceptable rates in 399°C (750°F) steam (1500 psi) as shown in Table III. Furthermore, more than twenty large, reactor-size, fuel tubes have been diffusion annealed at 890°C (1634°F) and corrosion tested for twenty-four hours in 343°C (650°F) water, then twenty-four hours in 399°C (750°F) steam (1500 psi). The satisfactory black tarnish films, which formed on these tubes, and the corrosion rates deduced from hydrogen evolution data have provided further evidence that the diffusion heat treatment does not impair the Zircaloy's corrosion resistance. These results are consistent with the results of a systematic study of the effect of heat treatment on the corrosion resistance of Zircaloy-2 indicating that 871°C (1600°F) with air cooling is one of the best heat treatments.[26]

One of the undesirable effects which may result from a diffusion heat treatment is an excessive increase in the concentration of uranium at or near the surface of the Zircaloy cladding. Large amounts of uranium might release fission products by recoil to the coolant thus giving false indications of fuel failure or raising the normal coolant activity so much that a fuel failure would be obscured.

Data have been obtained which showed that heat treatments which cause diffusion in excess of that necessary to prevent corrosion failure may also cause an increase in uranium concentration in the surface layers of the cladding.[22] By proper selection of cladding thickness and heat treatment, an increase in uranium content of the cladding near the surface can be prevented. Two such treatments, which also prevent corrosion failure, are shown in Table II. The data of this table provide evidence that the standard heat treatment (7 hours at 880°C) does not increase the uranium concentration in the outer 4 mils of 15-mil thick Zircaloy cladding.

The effect of the diffusion anneal on the corrosion rate of fuel elements during failure initiated at large defects in the cladding will be discussed in the following section.

4. Effectiveness in Full-size Tubes - The culmination of the development work was the successful heat treatment of full-size reactor tubes. This was accomplished by water quenching evacuated steel cans containing the fuel tubes after the 7 hours at 880°C standard heat treatment which was established during the previously described development program. Tubular sections from full-scale tubes were diffusion heat treated with 7-mil diameter defects drilled through the outer cladding but not into the core material. They survived several hundred hours' exposure to 343°C (650°F) water before the tests were terminated. Similar indication that the diffusion anneal effectively improved the cladding protection of full-size tubes was obtained from specimens made from sections of heat treated, full-size reactor tubes. These specimens were of necessity defected after the heat treatment. Proof that the defect reached the core material was obtained by examination of the corroded defect cross sections.

5. Explanation - A complete explanation for the protective action of the diffusion anneal has not been established. The evidence accumulated during the development studies indicates that bond strength, cladding stiffness, and cladding strength play major roles, while corrosion resistance of the core material plays a lesser role. As was previously mentioned, diffusion treatments which prevent corrosion failure increase both bond strength and cladding hardness; they may also strengthen the cladding in the 4 or 5-mil thick zone adjacent to the core. Further evidence of the importance of the cladding strength or stiffness is provided by the fact that thicker, hence stiffer, cladding increases the effectiveness of a diffusion anneal.

A strong bond and stiffer cladding may prevent the bulging and separation of cladding from core which occurs as corrosion progresses in rods which aren't diffusion heat treated. The area for direct core corrosion to occur is thus restricted to the base of the defect only. Further corrosion of the core material could be retarded if the initial corrosion product could be self-compacted enough to prevent direct attack of fresh core material by corrodant. Subsequent corrosion would occur by a process similar to diffusion through a sound oxide layer. The increased corrosion resistance of core material adjacent to the cladding may contribute to the formation of an oxide plug by forcing corrosion to progress inward more rapidly than adjacent to the bond line. The pear-shaped hole thus formed would redirect the forces exerted by the initial corrosion product so that they act to compact the corrosion product rather than to separate the core and the cladding. Evidence that the corrosion is restricted to the base of a plugged defect is shown in Fig. 2, which shows a cross section of a defect in a diffusion-annealed rod after 1100 hours in 343°C (650°F) water. The cladding is 30 mils thick. This particular defect was originally 40 mils deep and penetrated the core for 5 mils beyond the diffusion layer, thus exposing core material directly to corrosive attack.

The Canadians[8] have also attributed the beneficial effect of the diffusion anneal to the increased bond between the cladding and the core. They apparently place more emphasis, however, on the improved corrosion resistance of the portion of the diffusion zone formed by diffusion of zirconium into the core material.

Corrosion of Defected Clad Elements in Water

1. Philosophies - Much of the information discussed to this point has been concerned with the design philosophy whose goal is an absolutely reliable fuel element which won't corrode to contaminate the coolant. Perhaps the culmination of this approach is represented in the use of clad uranium oxide. Metallic fuels are more attractive on the basis of other considerations. Unfortunately, the closer a metallic fuel is made to approach the goal of 100% reliability, the higher its price becomes. Thus, it is not surprising that, as emphasis has shifted from technical feasibility to economic practicality, another design philosophy has developed. The goal of this new philosophy is to make the elements as reliable as is economically practical, but to be prepared for failure and design reactors capable of tolerating failure. This shift in emphasis has resulted in an increased interest in the manner in which a Zircaloy-clad fuel element with a core of uranium, or U - 2 w/o Zr, fails under conditions it might experience in a reactor. The magnitude and rate of progress of corrosion failure of such fuel under various conditions have been studied. Such data aid reactor designers in predicting whether failure can be detected in time to permit removal of the defective fuel before it is destroyed.

2. Progress of Corrosion During Failure

a. Comparison of Zircaloy-clad Cores of Uranium and U - 2 w/o Zr - As was indicated when the corrosion of bare unalloyed and alloyed uranium was discussed, the corrosion rate of the U - 2 w/o Zr was significantly less than that of the unalloyed uranium. This difference is more pronounced when one visually compares the progress of corrosion in defected rods with cores of unalloyed uranium and U - 2 w/o Zr. The specimens on the left side of Fig. 3 have unalloyed cores; those on the right have U - 2 w/o Zr cores. Those with U - 2 w/o Zr cores were exposed to 227°C (440°F) water for approximately four times longer than those with uranium cores and similar defects, yet corrosion has not progressed as far.

b. Effect of Diffusion Heat Treatment on Rods - The effect of a mild diffusion heat treatment on rods with cores of U - 2 ^{w}/o Zr in claddings with gross defects is illustrated in Fig. 4. The beneficial effect of the heat treatment is apparent even when the cladding defects were 40 mils diameter and 40 mils deep. In addition to reducing the severity of the failure at 277°C (530°F), the heat treatment modifies the nature of the failure. In as-extruded material, the cladding bulges and cracks unevenly, while failure progresses longitudinally. Heat treatment seems to localize the failure which develops with radial symmetry around the defect.

When the core was unalloyed uranium rather than U - 2 ^{w}/o Zr, little, if any, improvement in corrosion behavior was derived from the heat treatment. This difference in behavior can perhaps be explained if the Zircaloy cladding can extract carbon from the unalloyed core and form a zirconium-carbide layer,[27] which weakens the core-cladding bond. Since strengthening of the bond by the diffusion treatment is believed to be a major reason for the prevention of failure of Zircaloy-clad U - 2 ^{w}/o Zr fuel elements, a diffusion heat treatment of Zircaloy-clad unalloyed uranium might have the opposite effect to that desired if the carbon content of the uranium exceeded a minimum level by a sufficient amount.

c. Progress of Corrosion with Time - 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 and qualitative data on the progress of corrosion in water at three different temperatures and in steam at one temperature were obtained with sections from full-diameter tubes. Information was also obtained on the extent of corrosion during a simulated reactor cooling step following the initiation of shut down.

1. In Rods - The progress of corrosion in diffusion-annealed rods similar in all respects except for time subjected to corrosion is revealed by Fig. 5. These rods had U - 2 ^{w}/o Zr cores contained within 15-mil thick Zircaloy cladding in which gross defects were inserted. 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 results were obtained at both 277°C (530°F) and 299°C (570°F).

2. In Tubes - Tubular sections suitably prepared from full-size reactor fuel elements have been corrosion tested with gross cladding defects. The specimens used were made from sections of full-size fuel tubes with an OD of 2.060 inches and an ID of 1.467 inches; both inner and outer Zircaloy-2 cladding have a nominal thickness of 15 mils; fuel material is U - 2 ^{w}/o Zr. The progress of corrosion failure can be followed by using a wet test meter to periodically

measure the hydrogen evolved. The total amount of uranium converted to oxide can be calculated using a factor of 5 grams uranium/liter of hydrogen measured at room temperature.[6,7] A plot of such data for 299°C (570°F) is shown in Fig. 6 for as-extruded and diffusion-annealed tubes with gross cladding defects. Similar data has been obtained at 277°C (530°F) and 349°C (660°F). Such plots can be used to deduce the extent of corrosion during specified intervals following the achievement of a corrosion level capable of signaling failure. Suppose, for example, the reactor is designed to detect failure after 2.5 or 5 grams of uranium have been oxidized. The plots for the proper coolant temperatures may then be read to establish the time, $T_{s}(2.5)$ or $T_{s}(5)$, at which 2.5 or 5 grams have been oxidized. The amount of uranium converted to oxide during intervals following the respective T_s can then be obtained from the plot. Data obtained in this way are given in Table IV. Such data show that at 277°C (530°F) and 299°C (570°F) the diffusion heat treatment provides an added margin of safety. For various intervals after the detection of failure, heat-treated material undergoes less attack than as-extruded material. These results also serve to 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.

A visual comparison of the progress of corrosion in as-extruded and heat-treated tubular samples corroded at 349°C (660°F) is shown in Fig. 7. The marked change in the nature of attack as a result of heat treatment is perhaps even more striking than for small specimens (see Fig. 4) despite the higher test temperature. 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 of tubes 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 fuel tube, or to render removal of the process tube difficult. The rupture of an as-extruded tube may cause blockage of coolant channels and hinder the removal of the failed tube unless failure is arrested at an early stage.

3. Corrosion Progress Under Reactor Shut Down Conditions - It is possible to define the pattern of cooling for a reactor after shut down.[28,29] This cooling pattern can then be used to terminate corrosion tests in which the initiation of shut down is simulated at specified time intervals after corrosion has reached a set reference level. Results for different intervals then indicate how long after the reference level is attained the reactor operator can afford to wait to initiate shut down. Tubular sections made from full-size

fuel elements have been given gross cladding defects and tested with simulated reactor shut down. The results of some tests are summarized in Table V. The corrosion progress was once again followed by measuring hydrogen evolved. Photographs of some of the corroded specimens are shown in Figs. 8 through 11.

These tests show that the in-pile failure of a power tube in 350°C water is not likely to have disastrous consequences. A delay of 60 minutes between the attainment of 5 grams of total uranium corrosion level and the initiation of shut down does not lead to such severe damage to the tube as to introduce serious difficulties in removal of the tube from the reactor. The total uranium corroded at the end of reactor shut down is not likely to exceed 300 grams and is, in fact, likely to be closer to 100 grams.

These tests provided further evidence that the pattern of core consumption and clad damage is changed by heat treatment and that the cooling rate following heat treatment should be fairly rapid. Corrosion of as-extruded material proceeds longitudinally from the defect; penetration of the core is so shallow that distortion of the inner cladding is unusual. The outer cladding undergoes bulging and lengthwise splitting [see Fig. 7(a) and 9(a)]. The corrosion of diffusion heat-treated [7 hours at 880°C (1616°F)] or less severely heat-treated [1/2 hour at 800°C (1472°F)] material is more symmetrical about the defect [see Fig. 7(b) and 9(b)]. Inward penetration seems to be favored, reaching the inner cladding and distorting or even cracking it.

The relatively poor performance of samples 12-1-D and 12-1-E is believed to have resulted from a slow cooling rate after heat treatment of the full tube.

4. Tests in 350°C (662°F) Steam - For prediction of in-pile corrosion behavior in some reactor situations, information is needed on the behavior of a defected fuel element in steam. The pressure in the coolant system may be below 800 psi, corresponding to a dew point below 300°C (572°F). Therefore, steam, not water, can be in contact with the fuel elements at 350°C (662°F), the temperature selected for the shut down tests. A comparison between corrosion in water and steam at 350°C (662°F) was obtained with the assistance of personnel at Argonne National Laboratory, who made available a flowing steam autoclave. Hydrogen in the effluent could be measured with a wet test meter. Assuming that hydrogen evolution reflects the progress of corrosion,[11] comparisons can be made on the basis of hydrogen evolution as a function of time after attainment of the reference level for total uranium corrosion. Corrosion data from such steam tests of tubular specimens similar to those previously used is shown in Table VI. For the first 35 minutes after attainment of the reference level (5 grams of uranium corroded or one liter hydrogen evolved), the corrosion for three out of four tests in steam was about the same as that in water. Sixty minutes after the reference level was reached, the corrosion in steam was two to four times that in water.

The physical appearance of the steam sample was a reasonable extension of the appearance of water specimens whose tests had been completed with less hydrogen generation. The outer cladding had split and rolled back. The inner cladding also bulged and started to rupture. The Zircaloy near the rupture sparked on exposure to air and had become brittle, indicating that hydride formation probably played a more significant part in the steam tests than in the usual water tests.

In summary, the corrosion of a defected, diffusion heat-treated, power fuel tube is more rapid in 350°C (662°F) steam than in 350°C water, perhaps because of hydride formation in the former. The pattern of failure is similar in the two media. In either case, in-pile failure of a power tube is not likely to have disastrous consequences. The high flow rate of the coolant in an actual reactor may prevent the hydrogen concentration near the exposed core material from reaching the levels attained in these tests.

5. Effect of Hydrogen - The effect of hydrogen must be considered in the evaluation of the corrosion of uranium and its alloys in a water-cooled reactor. Hydrogen may arise from several sources: reaction of uranium with water; radiolytic decomposition of the water; and deliberate addition to suppress corrosion of other reactor components. Various tests of tubular specimens at 299°C (570°F) indicated a relative lack of sensitivity of corrosion to extremes of hydrogen concentration in the autoclaves. It has been concluded that at this temperature higher hydrogen concentrations do not significantly affect the corrosion of a power tube.

6. Effect of Aging - As was previously mentioned, the addition of 2 $^{w}/o$ Zr reduces the corrosion rate of uranium by a factor of from three to ten. Since the solid solubility of zirconium in uranium is less than 1 $^{a}/o$, i.e., 0.4 $^{w}/o$, the possibility has to be considered that the observed improvement in U - 2 $^{w}/o$ Zr is due to supersaturation by zirconium and that an aging treatment precipitating this excess zirconium can lead to a loss of a metastable 2 $^{w}/o$ Zr alloy's advantage. Thermal conditions conducive to aging may occur during reactor operation. Evidence has been obtained that corrosion rate of the diffusion-annealed 2 $^{w}/o$ Zr alloy is insensitive to subsequent thermal aging at lower temperatures. Specimens of the bare alloy aged at 450°C (842°F) to 460°C (960°F) for twenty-four hours or 600°C (1112°F) for two hours after the standard diffusion anneal corroded at essentially the same rate as those which were either as-extruded or diffusion annealed. Furthermore, a number of other bare alloy specimens cooled in several different ways after the standard diffusion anneal corroded at essentially the same rate.

SUMMARY AND CONCLUSIONS

If uranium or its high alloys are protected by a corrosion-resistant cladding, they may be used as fuels for water-cooled nuclear reactors. Acceptance of such elements by reactor designers will increase the prospects for fueling these reactors with natural uranium. A large amount of effort has demonstrated that cladding applied by coextrusion is very trustworthy and that failure, if it does occur, can be minimized and need not be disastrous. Additional effort has established techniques for testing the integrity of the cladding and for improving its reliability. Fuel elements made of U - 2 $^{w}/o$ Zr coextruded with Zircaloy-2 cladding have been the subjects of considerable development work whose end view is to provide tubular fuel elements for a heavy water-cooled reactor.

The protection provided by Zircaloy-2 cladding for U - 2 $^{w}/o$ Zr is increased by application of an interdiffusion heat treatment. This treatment prevents failure of fuel elements with tiny or subtle defects such as are simulated by a cylindrical hole of 7 mils diameter. The protective action appears to be retained after irradiation to low burnup. A diffusion heat treatment is most effective when the cladding is at least 15 mils thick. Various temperatures from 800 to 1100°C have been successfully used; the lower temperatures require longer times to be effective. 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. The optimum rate of cooling has not been established.

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.

The standard heat treatment was not found to produce any adverse effects on other properties of the Zircaloy-2 cladding.

Quantitative data on the progress of corrosion have been obtained by continual measurement of the hydrogen evolved. These data on tubular samples with gross cylindrical defects tested in water at 277°C (530°F) or 299°C (570°F) have shown that heat treatment leads to slower corrosion in various periods after corrosion has reached the level necessary to provide a signal of failure in the reactor situation under consideration.

Tests of tubular sections with gross defects have shown that in-pile failure of an element in water or steam at 350°C (662°F) is not likely to lead to disastrous consequences. When the core alloy is U - 2 $^{w}/o$ Zr, a delay of 60 minutes between attainment of the 5-gram uranium corrosion level in water and the initiation of shut down may permit discharge of several hundred grams of uranium into the coolant stream. The most likely uranium release is near 100 grams. Corrosion in steam may be more rapid by a factor of about 2 to 4. In both media, the physical damage to a power tube is not expected to introduce serious difficulties in its removal from the reactor. Although a diffusion heat treatment provides added protection against subtle defects, the lowest corrosion rate in these 350°C tests has been observed in as-extruded material.

In such material, corrosion and damage to the cladding proceed longitudinally. Even a mild heat treatment changes the pattern of corrosion to one that is more symmetrical about the defect and produces a greater tendency to consume the core inward, reaching and distorting the inner cladding.

It has not been possible to find differences between the behavior of different lots of tubes. Increasing hydrogen concentrations have no noticeable effect on corrosion at 300°C. Aging at 450°C or heat treatment at 600°C does not undo the advantage of the U - 2 $^{w}/o$ Zr alloy over unalloyed uranium. Gross defect tests of tube sections with unalloyed cores are needed for confirmation of the previously demonstrated advantage of alloying.

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Table I

Corrosion Rates of Uranium-Zirconium Alloys

($g/cm^2/hr$)

Temp.		Unalloyed U (1,2,3,4)	U-2 w/o Zr (1,3)	U-5 w/o Zr (1,5)	U-7.5 w/o Zr (1)	U-10 w/o Zr (1,5)	U-15 w/o Zr (1)
°C	°F						
100	212	0.003	0.001-0.003	0.001	0.0007	0.0003-0.001	
150	302	0.036	0.036				
200	392	0.23	0.23	0.16		0.06	
227	440	0.7-2.0	0.4-0.6	0.6	0.5	0.36	
250	482	2	1.0				
260	500	1.4-3.0	1.0-1.1	1.0	1.0	0.5-0.7	0.5
277	530		1.3-1.5				
299	570		2.0-3.0				
300	572	4.4-4.8	1.7-3.7	2.1-2.8			
316	600	5-10	3.0-5.0	5.0		2.0	
343	650		6.0-7.0	6.0-10.0	2.5	5.0-(7)	

(1) NMI-4364.
(2) BMI-998.
(3) CRDC-646.
(4) BMI-1273.
(5) BMI-1156.

Table II

Increases* in Uranium Concentration (ppm) in Successive Layers of Zircaloy-2 from As-extruded and Heat-treated Rod Samples**

Core: U - 2 $^{w}/_{o}$ Zr (nominal)

Distance From Surface of 15 Mils Thick Cladding	As-extruded		9 hr/880°C		8 hr/900°C	
	Min	Max	Min	Max	Min	Max
Outermost mil	8.87	889.0	1.88	20.13	1.19	12.32
Second	0.3	0.92	1.92	5.81	2.11	6.49
Third	0.0	6.79	0.33	2.82	1.46	1.76
Fourth	0.08	0.33	0.26	1.37	0.91	2.15
Fifth	0.73	3.43	1.15	1.32	0.44	3.15
Combined sample from outermost four mils	0.80	1.99	0.98	1.04	1.39	18.84

* Value presented represents amount of uranium in excess of that found in a reference sample such as the Zircaloy stock used to make specimens.

** Samples obtained from two extruded rods.

Table III

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 From End Seals	Clad Rods	Zircaloy Cropped From End Seals
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

Table IV

Corrosion Progress* in Tubular Fuel Element Sections in 277°C (530°F) Water

Defect: 40 mils diameter; 40 mils depth
Cladding: 15 mils thick Zircaloy-2
Core: U - 2 $^{w}/o$ Zr (nominal)

Specimen	Heat Treatment	$T_{s(2.5)}$** (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	None	7.75	0.5	1.5	3.7	15.3				12.25	4.5
3-10-B		6.30	13.3							7.75	1.5
3-10-C		10.50	1.0	9.5	22.0	38.0				15.25	4.8
3-10-D		4.80	13.8	52.8	118.8					8.00	3.2
7-H2-A	7 hr/880°C rapid cool	6.00	2.0	3.5	6.0	8.5	15.0	23.5		14.00	8.0
7-H2-B		6.00	1.0	2.0	3.5	5.0	13.0	19.0		19.75	13.8
7-H2-C		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	None	10.50	8.8							12.25	1.8
3-10-B		6.65	21.1							7.75	1.1
3-10-C		11.75	9.5	23.5	39.1					15.25	3.5
3-10-D		5.10	21.1	70.0	150.0					8.00	2.9
7-H2-A	7 hr/880°C rapid cool	7.40	2.0	4.0	7.0	10.3	18.5			14.00	6.6
7-H2-B		8.40	1.8	3.0	7.5	12.0	17.5	23.8		19.75	11.4
7-H2-C		6.75	2.3	4.8	7.5	10.5	17.5	25.0	66.3	25.50	18.8

* Followed by measurement of hydrogen evolution using wet test meter.

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

*** T_t = termination time of test.

Table V

Corrosion Tests of Tubular Fuel Element Sections Under Simulated Reactor Shutdown Conditions in 350°C Water

Defect: Diameter, 25 mils; Depth, 40 mils
Cladding: 15 mils Zircaloy-2
Core: U - 2 w/o Zr (nominal)

Sample No.	History	Time at Temp. to 1 Liter H_2 (min)	Total H_2 Collection at Shutdown at Time t After 1 Liter		Residual H_2 (liters)*	Total H_2 (liters)*	Measured Weight Loss (grams)
			t (min)	Total H_2 Time t (liters)			
17-C	As-extruded	45	30			4.99	
21-2A-2		70	60	5.78	0.92	6.70	39
13-2-C	SDA** cooled rapidly, by quenching container in water	60	35			3.84	
13-4-B		45	60			7.53	
21-B-5		30	60	15.04	4.70	19.74	106
21-B-7		15	60	27.64	(23.06)	(50.70)	301
25(15 - 20)		25	60	18.87	20.20	39.07	240
25(21-1/4 - 26-1/4)		35	60	11.07	Not measured		87
17-G-1	SDA** cooled more slowly by cooling container in air	30	60	16.68	10.85	27.53	
17-G-2		50	60	6.98	2.17	9.15	56
12-1-D	SDA** cooling rate presumably slow because of Ta wrapping	25	2			4.84	
12-1-E		40	25			18.92	

(Cont'd. on next page)

Table V (Cont'd.)

Sample No.	History	Time at Temp. to 1 Liter H_2 (min)	Total H_2 Collection at Shutdown at Time t After 1 Liter		Residual H_2 (liters)*	Total H_2 (liters)*	Measured Weight Loss (grams)
			t (min)	Total H_2 Time t (liters)			
12-2-F	SDA** slow cool in Ta wrapping plus 1 hr/880°C; rapid cooling	125	60	5.41	2.36	7.77	53.8
13-4-A	SDA** rapid cooling plus 1 hr/880°C; rapid cooling	60	60	5.59	0	5.59	32.7
17-K	SDA** extremely rapid cooling; specimen quenched by water	65***	60	10.34		(20)	97
21-2A-1B	1/2 hr/800°C; rapid cooling	30	60	19.04	14.67	33.71	177

* Residual hydrogen was generated during simulated shutdown and measured at room temperature. Residual hydrogen or total hydrogen does not include hydrogen left in autoclave at atmospheric pressure (estimated at 1.5 liters).

** SDA = Standard diffusion anneal, i.e., 7 hr/880°C in evacuated container.

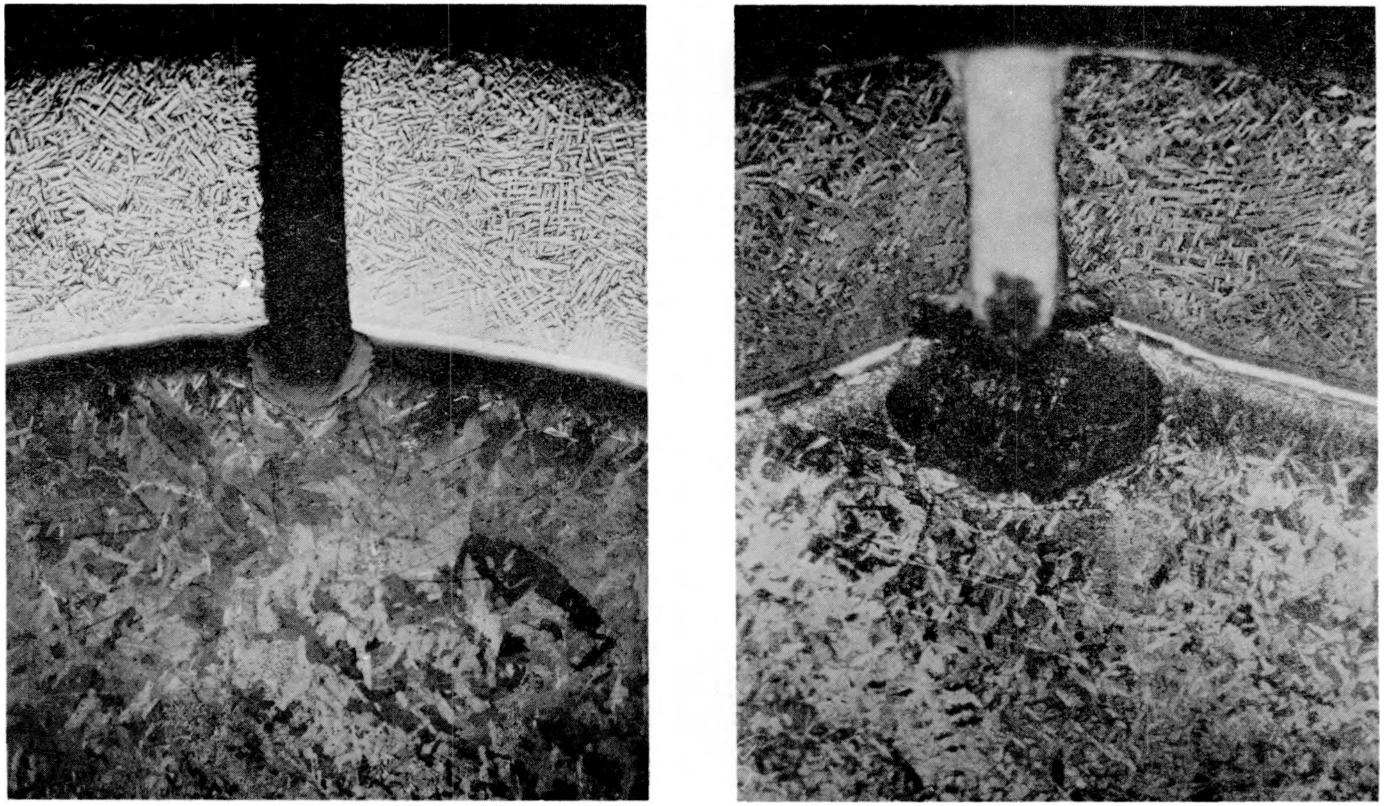
*** Time at temperature during second test. Only 0.1 liter H_2 was evolved during prior 6 hours at temperature in first run.

Table VI

Comparative Hydrogen Evolution in 350°C Steam and 350°C Water Tests

Sample No.	Medium	Time at Temp. to 1 Liter Signal (min)	Total H_2 Evolution, Liters, at Time t After 1 Liter			
			t = 35 min	t = 60 min	t = 75 min	t = 90 min
13-2-C	Water	60	3.84*			
13-4-B	Water	45		7.53*		
13-4-C	Steam	100	4.91	11.54	18.28	25.44
13-4-E	Steam	100	3.30	12.16	17.69	26.50
13-4-F	Steam	110	3.66	13.00	20.91	
13-4-D	Steam	120	11.98	23.12		

* H_2 was measured after autoclave cooled and therefore includes H_2 generated during simulated shutdown from 350°C.

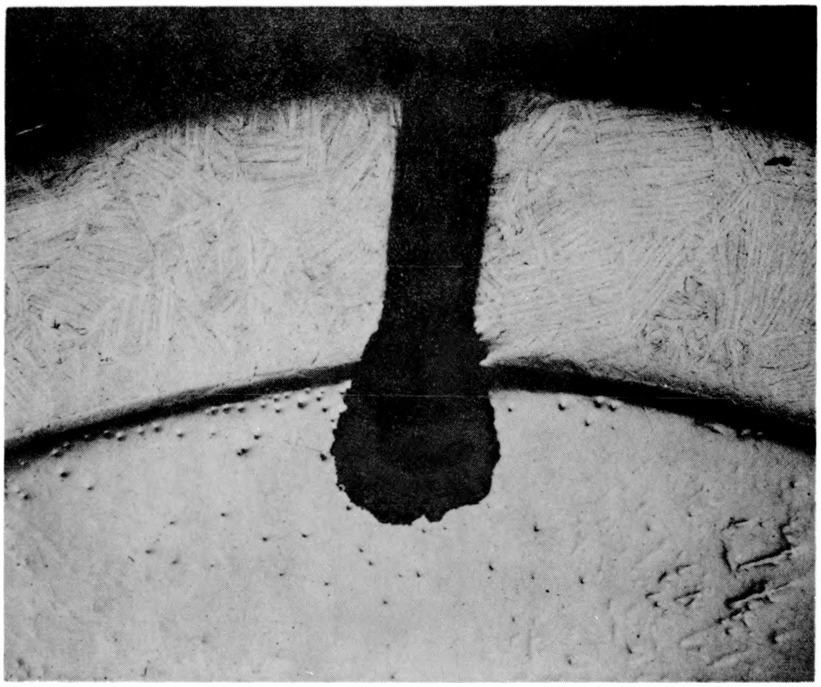


128 hr in 600°F Water

1073 hr in 600°F Water
2700 hr in 750°F Steam

Fig. 1

Corrosion of Diffusion-Annealed Specimens of U - 2 $^{w}/o$ Zr Clad with Zircaloy-2. 7-mil Diameter Defect Inserted Before Heat Treatment. Polarized Light. 50X Magnification.

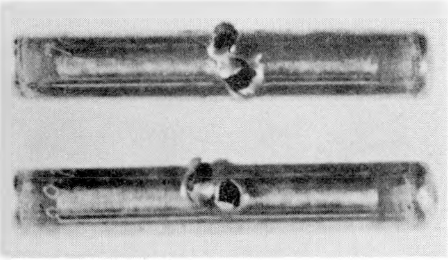


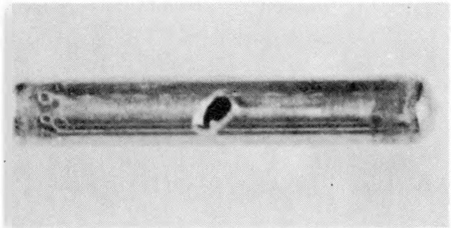
50X Bt. Lt. A-1108-(a)

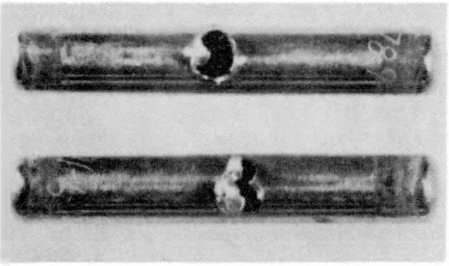
Fig. 2

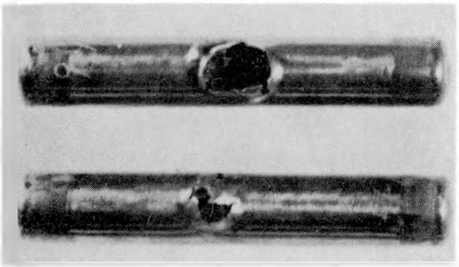
Corrosion at Base of 7-mil Diameter Defect Drilled 10 Mils Into the Core Below 30-mil Thick Cladding.

Diffusion-annealed for 2 hr at 1050°C. Corrosion tested for 1100 hr in 343°C (650°F) water.









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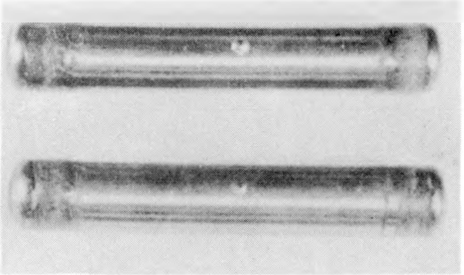

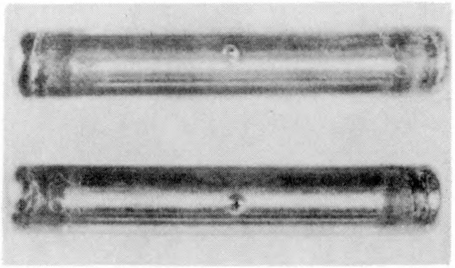

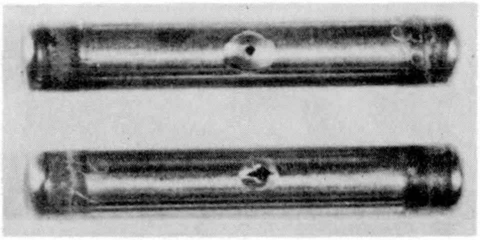

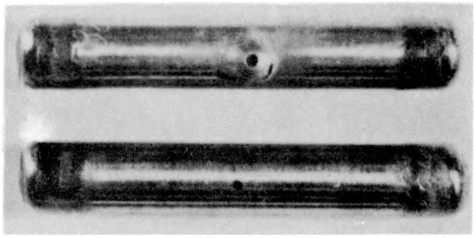

RF-5392

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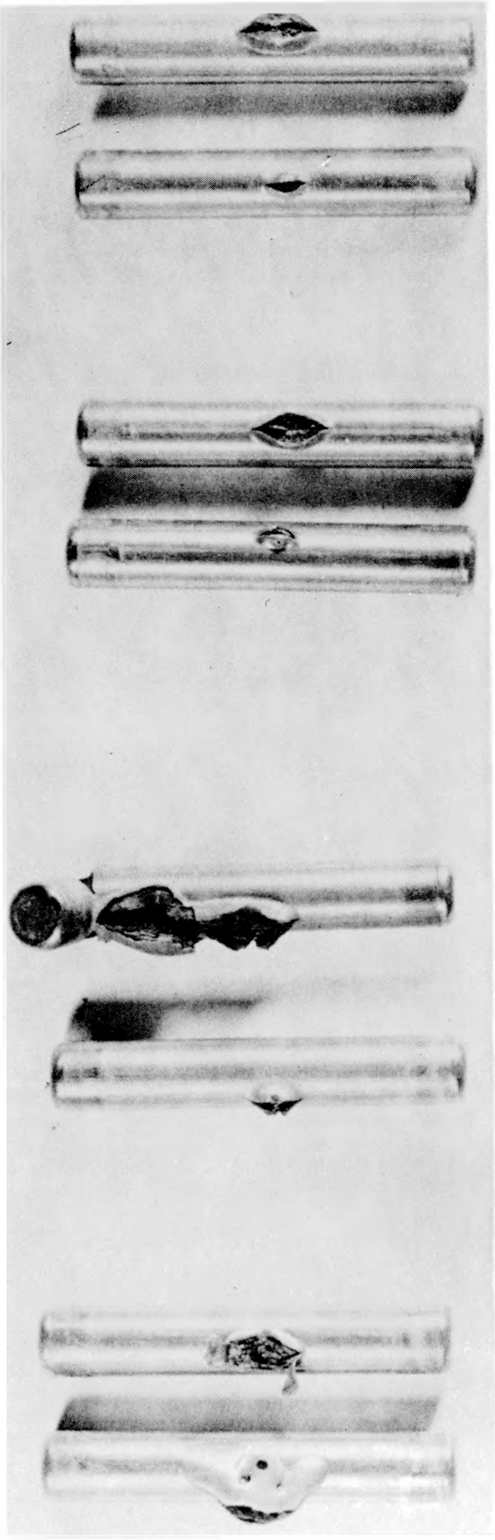

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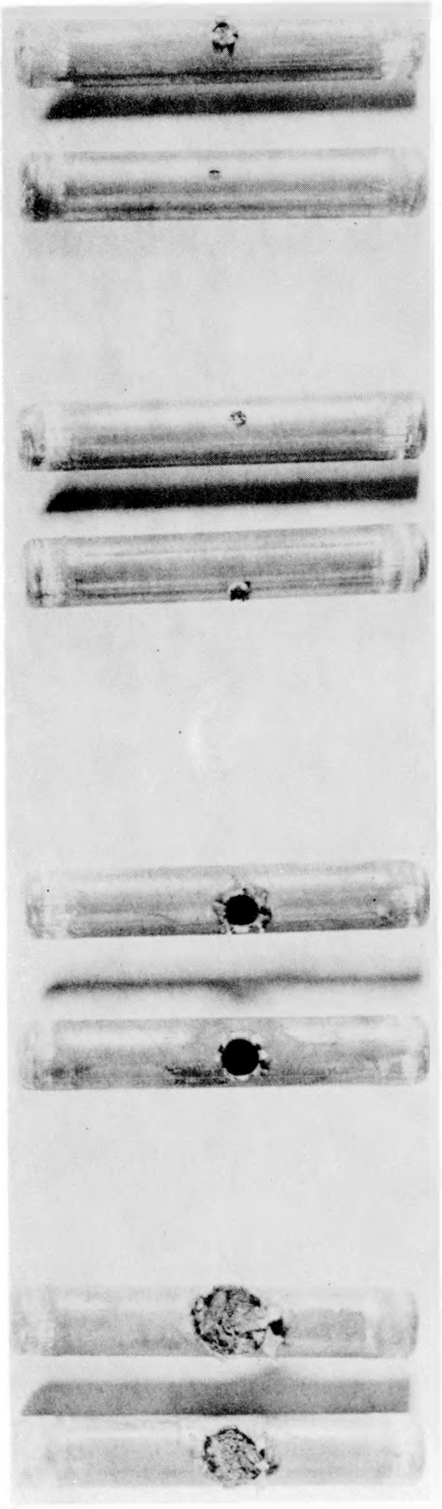

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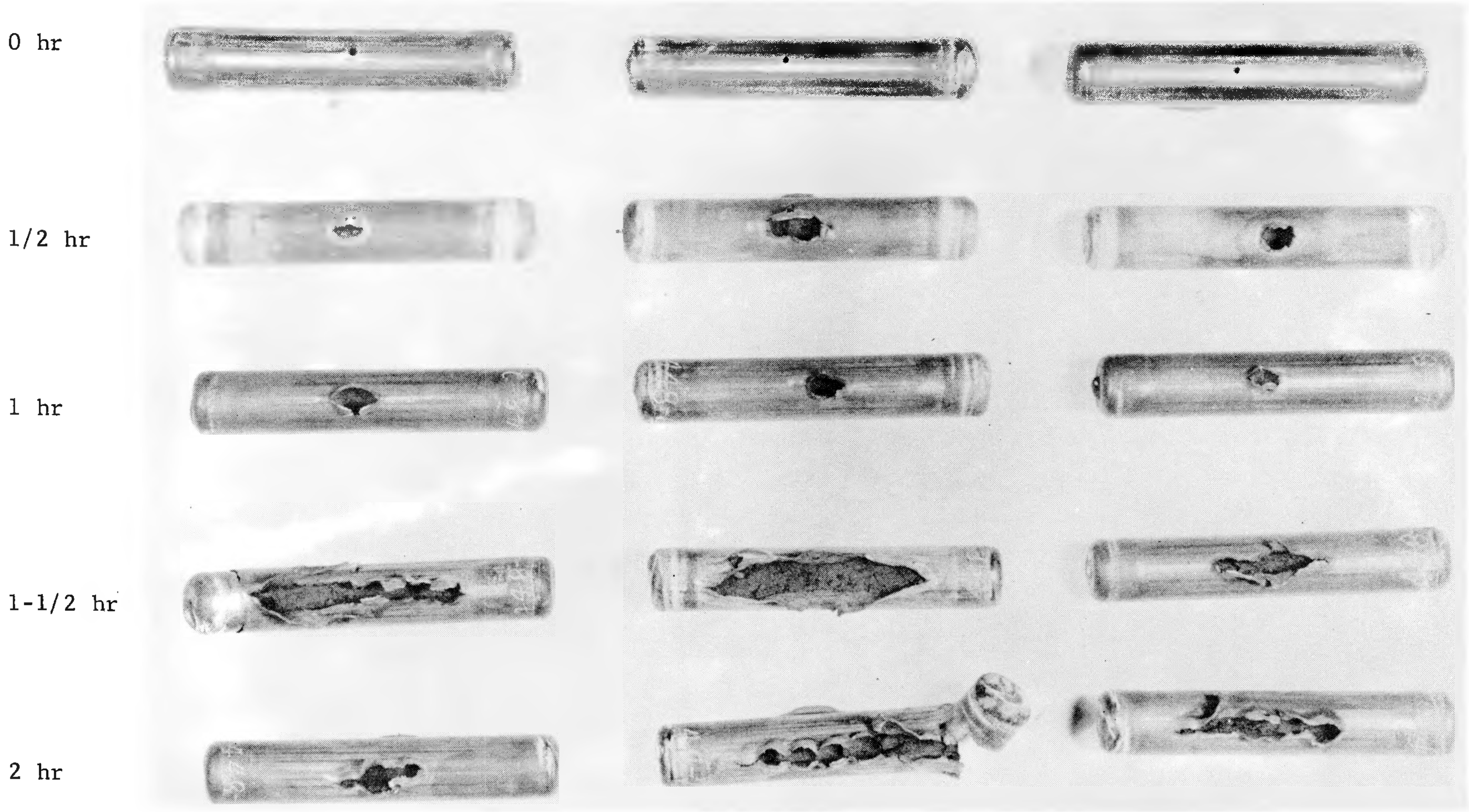


<u>Fig. 5</u> - Progress of Corrosion at 299°C (570°F).
Defect Diameter - 40 mils.
Depth - 40 mils.
Nominal Length of Each Sample - 2 inches.
Zero Hour = autoclave containing specimen was cooled immediately after attaining test temperature.

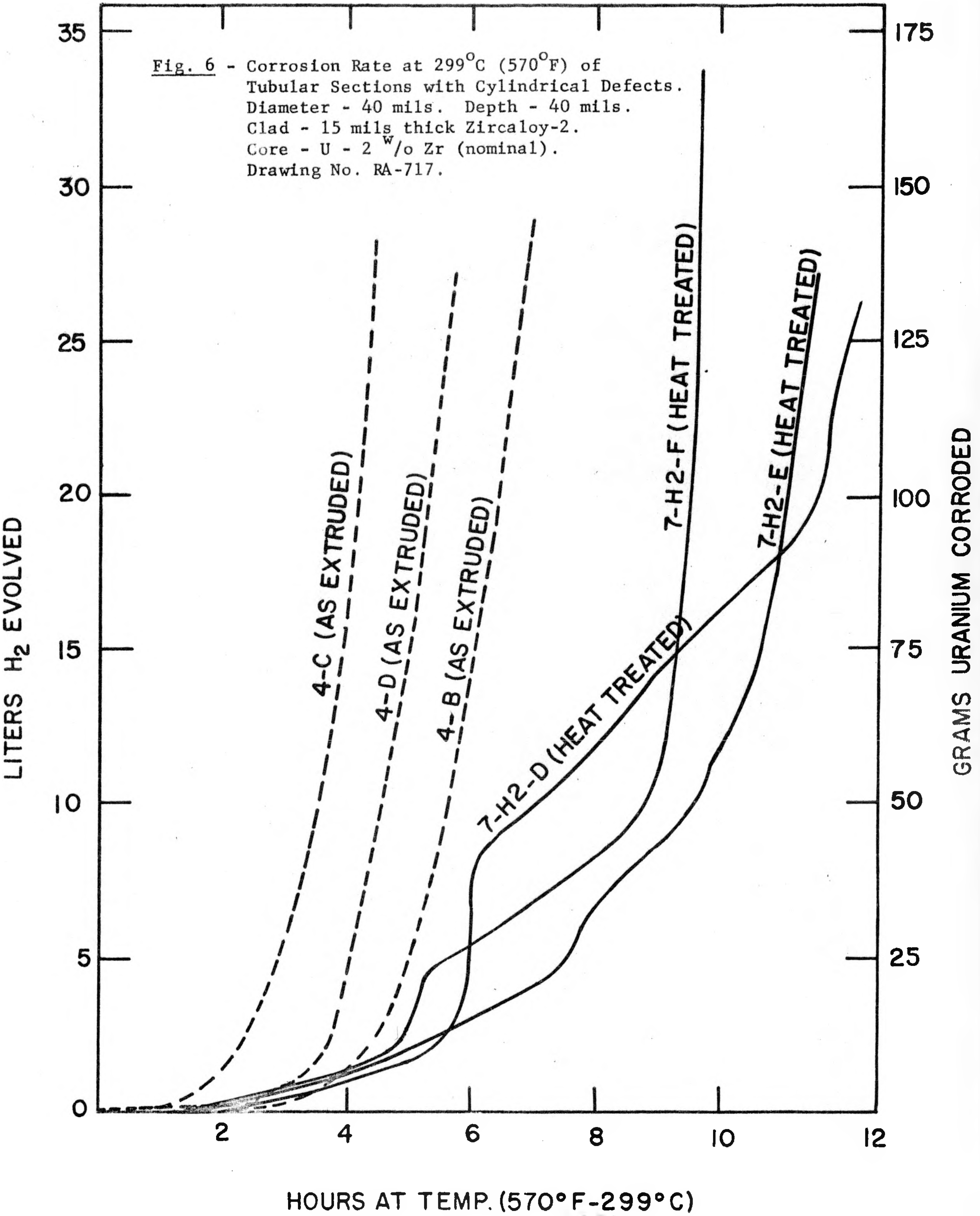
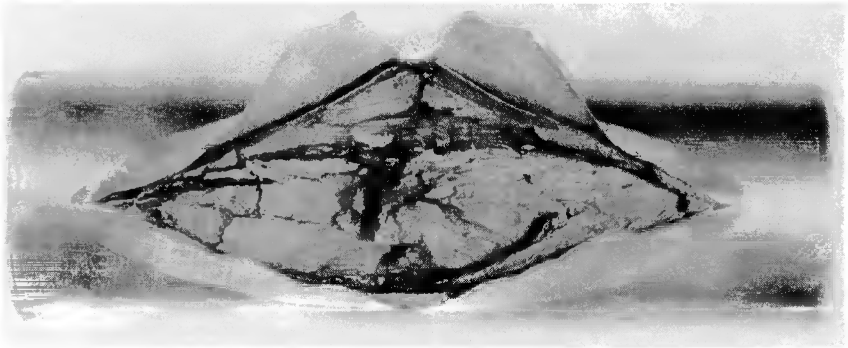
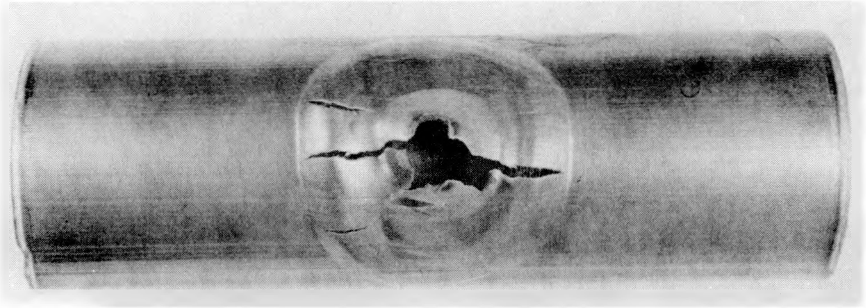


Fig. 6 - Corrosion Rate at 299°C (570°F) of Tubular Sections with Cylindrical Defects. Diameter - 40 mils. Depth - 40 mils. Clad - 15 mils thick Zircaloy-2. Core - U - 2 w/o Zr (nominal). Drawing No. RA-717.



(a) RF-5737

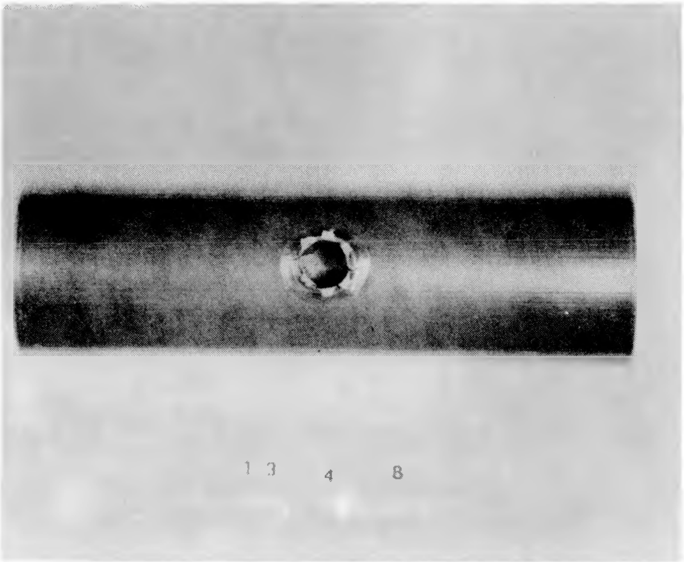
As-extruded
4-A
3 hr at 349°C



(b) RF-5738

Heat Treated
7 hr/880°C - Air-cooled
12-2-D
3/4 hr at 349°C

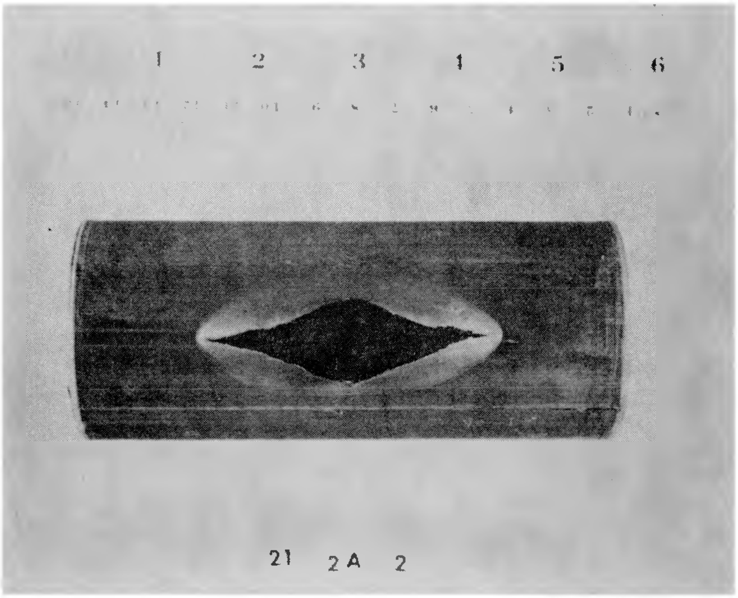
Fig. 7 - Tube Sections Tested in Water at 349°C (660°F) Original diameter is 2 inches.



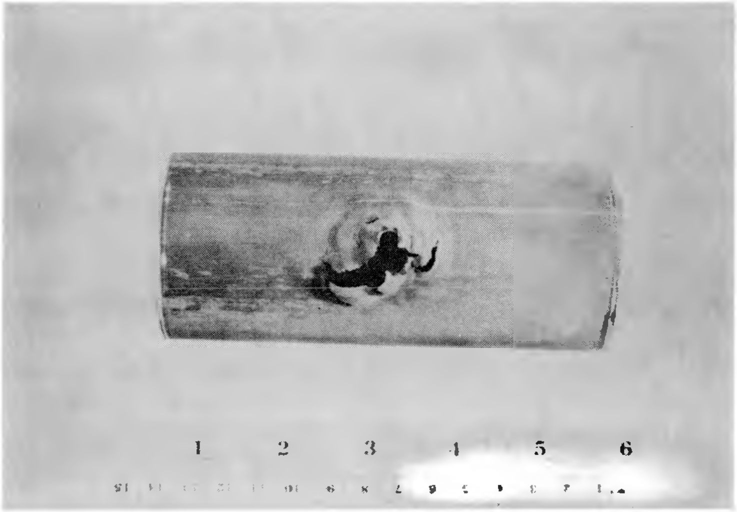
13-4-B RF-6053

Fig. 8

Sample 13-4-B from Heat-treated Tube Tested in Water at 349°C (660°F). See Table V for corrosion data. Original diameter is 2 inches.



(a) 21-2A-2 RF-6365

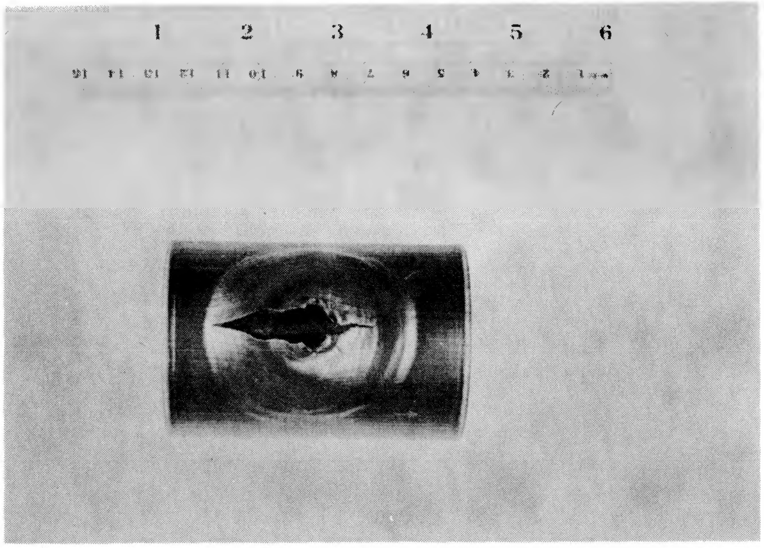


(b) 21-2A-1B RF-6403

Fig. 9 - Sections from Tube 21 with Different Thermal Histories.

(a) 21-2A-2 - as-extruded.
(b) 21-2A-1B - special heat treatment: 30 min at 800°C.

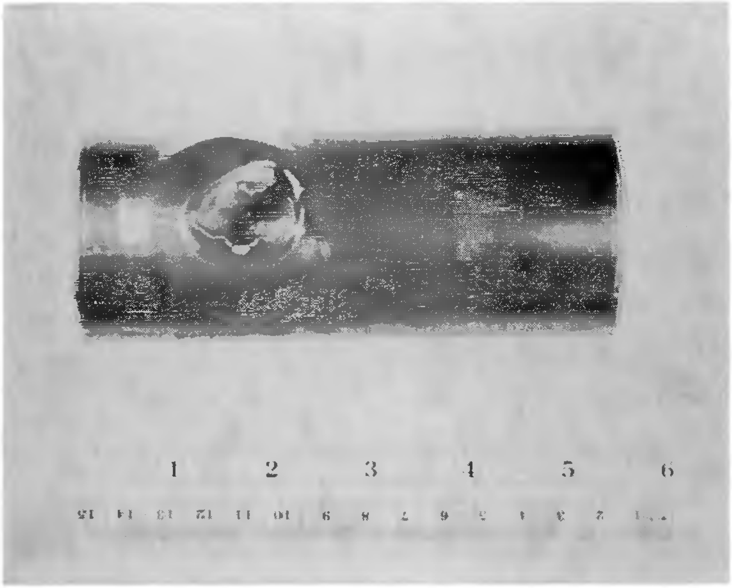
Tested in water at 349°C (660°F). See Table V for corrosion data. Original diameter is 2 inches.



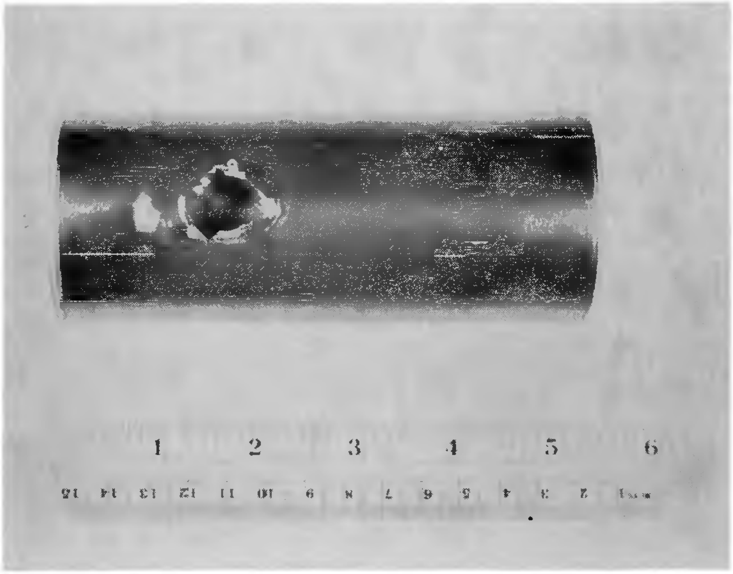
21-B-7 RF-6374

Fig. 10

Section from Tube Subjected to Standard Diffusion Heat Treatment (7 hr at 880°C) Tested in Water at 349°C (660°F). See Table V for corrosion data. Original diameter is 2 inches.



(a) 25(15 - 20) RF-6483



(b) 25(21-1/4 - 26-1/4) RF-6484

Fig. 11 - Sections from Heat-treated tube.

(a) Specimen 25(15 - 20).
(b) Specimen 25(21-1/4 - 26-1/4).

See Table V for corrosion data.
Original diameter is 2 inches.