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INTERIM REPORT: DEFECT TESTING OF COEXTRUDED
URANIUM-ZIRCALOY-II CLAD FUEL MATERIAL IN A
300 C OUT-OF-REACTOR RECIRCULATING WATER LOOP.

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I. INTRODUCTION

A major problem in the development of a pressurized water reactor coolant system for the NPR is the rupture performance of the fuel elements. As water temperatures are increased to 300 C, uranium corrosion rates increase rapidly. Swelling of the uranium fuel by corrosion could cause the process tube to burst or reduce the tube cooling water flow below acceptable limits. The desirability of slow cooling of the water to avoid thermal shocks to the reactor piping after a rupture is detected further complicates discharge and decontamination problems as fuel will continue to corrode with attendant fuel element damage during the cooling period.

Coextruded uranium-zircaloy-2 clad fuel elements are scheduled for use in the NPR. The rupture behavior of this type fuel material after heat treatment was studied in ELMO-4, an out-of-reactor recirculating water loop. Several types of initial defects were studied including pinhole and simulated cleavage. Most of the work consisted of measuring weight loss and physical dimension changes after various lengths of exposure in 300 C water. This report presents the results of the tests.

II. SUMMARY AND CONCLUSIONS

A series of tests were performed in ELMO-4, an out-of-reactor recirculating water loop, to determine the rupture behavior of co-extruded uranium zircaloy-2 clad fuel materials. All the tests were run at 300 C with water velocities of 7-20 fps and a system pressure of 1500 psi. Fuel materials with five different heat treatment histories and with different types of defects were tested to determine their rupture behaviors. The five conditions were (1) as-extruded material as received from Nuclear Metals, Inc., (2) beta heat-treated and water quenched, (3) beta heat-treated and air-cooled, (4) beta heat-treated, isothermally treated at 600 C and air-cooled, and (5) beta heat-treated and furnace cooled (vacuum). Both pinhole and cleavage type defects were studied.

The most desirable fuel material condition was beta heat treated, followed by isothermal treatment at 600 C and air cooling. A pinhole type defect in rods in this condition consistently produced only a single raised and torn mound about 3/8" in diameter and 0.13 inches high even after exposures of four hours. The uranium release rate was about 0.25 grams per minute after an initial 40 minute incubation period. This compares to a calculated 6.25 gram per minute uranium release rate from an equivalent sized bare uranium rod exposed for 1 hour at 280 C in ELMO 4. (1) Simulated cleavage type defects produced very minor swelling of the cladding even though large amounts of uranium were exposed and released. Specimens in some of the other conditions produced corrosion rates much greater than the isothermally treated pieces. Uranium release rates from rods in other conditions with a pinhole type defect were as great as 0.7 grams per minute. Also, rods in the other conditions usually displayed a great deal of swelling and splitting of the zircaloy-2 cladding. Average height of the defect was 0.25 inches after three hours exposure, with a maximum observed height of 0.31 inches. Swelling of the rods usually progressed along the longitudinal axis rather than radially.

Tubular fuel specimens defected with a pinhole on the outside surface ruptured almost entirely on the outside surface. The only exceptions were those which

(1) Hayden, K.D., Summary Report of Fuel Element Rupture Tests in High Temperature High Pressure Recirculating Water Systems, HW-48836, March 1, 1957. (Confidential)

failed in the unbonded end cap area and then only minor swelling was observed on the inside surface. Diameter increases of the tubes after failure were consistent with the rods and averaged about 0.15 inches for the isothermal treated pieces and 0.25 inches for the other pieces. One of the as-extruded tubular elements which was swept out of the 2.1" i.d. KER size process tube and into a 3" pipe, where consequently there was a large amount of space for expansion, had a maximum diameter of 2.44 inches, increased from the original diameter of 1.8 inches, after four hours exposure. This was the largest swelling of any of the elements tested.

The manner in which the coextruded fuel material is cooled during or following beta heat treatment appears to be a dominant factor in its subsequent failure behavior. The bond strength (as determined with a shear specimen) of the best behaved material is not the maximum attainable, and the possibility exists that some mechanical condition or structural feature in these bonds which alters the bond strength is also responsible for the decrease in corrosion rate and clad deformation during failure. Anomalous behaviors of some water-quenched specimens suggests that a heat treatment equivalent to or superior in behavior to the isothermal treatment may be developed.

Since the rupture experiments were conducted in an out-of-reactor test loop they may be only indicative of in-reactor ruptures. Bond damage during irradiation and heat evolution of the fuel material during the failure process might increase corrosion rates and clad deformations. Experiments to determine the effect of heat generation on the rate of rupture are currently in progress in another loop and will help to determine what correction factors need be applied to the ELMO-4 test results. (2)

III. DISCUSSION

A. The Problem

The rapid reaction rate between uranium and water at 300 C coolant temperature requires that the fuel element exhibit a vastly improved rupture behavior over the massive type elements presently in use at Hanford. (3) Coextruded Uranium-Zr-2 clad fuel material has shown in preliminary studies that the required improvement in rupture behavior may be attainable.

The rupture behavior of coextruded Uranium Zr-2 clad fuel materials needs to be characterized before fuel element specifications for the NPR can be established. Consequently, a detailed testing program has been initiated within Reactor and Fuels Research & Development Operation to evaluate the various parameters which may influence the fuel corrosion rate and the behavior of the fuel shape during failure.

This document reports the results of tests made in a dynamic, high-temperature, high pressure facility for some of the parameters of coextruded uranium-Zr-2 clad fuel materials. The parameters explored during the testing were:

(2) Hayden, K.D., Coextruded Uranium and Zircaloy-2 Rupture Test With Internal Heat Generation in a 300°C Out-of-Reactor Water Loop, HW-60775, June 1959,

(3) Op. Cit. Hayden, K.D., HW-48836.

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1. Fuel material geometry and clad thickness
 - a. 0.6" dia. x 0.030" clad rod
 - b. 1.82" O.D. x 1.45" I.D. x 0.020" clad tube
 - c. 1.78" O.D. x 1.38" I.D. x 0.030" clad tube
2. Nature of defect
 - a. Subtle - 0.025" dia. hole through bond and clad
 0.025" dia. hole in unbonded end cap zone
 - b. Severe - shallow full length and partial length slits
 - deep partial length slits
3. Temperature and pressure
300 C and 1500 psi
4. Heat treatment
Uranium beta treatment as it affects bond strength, diffusion thickness and structure.

B. ELMD-4 Loop Facility

The main requirement for establishing in-reactor failure behavior in an out-of-reactor system is that the tests provide complete or almost complete simulation of reactor operating conditions. Conditions that can be duplicated in an out-of-reactor system are water temperature, flow, pressure, and size and nature of the sample. Heat generation can be accomplished with special facilities. Neutron effects are lacking and can be evaluated only with in-reactor systems. The ELMD-4 loop has been designed and constructed to provide all the requirements except neutron and thermal flux conditions at operating conditions of 300 C, 1500 psi and 50 gpm flow. The fuel elements or assemblies to be tested are placed inside KER size Zircaloy-2 process tubes to simulate KER fuel element and tube configuration. Valves to isolate the test section are incorporated in the loop so the water can be heated to the required temperature before it is allowed to flow over the fuel elements. A heat exchanger in the system is used to cool the loop water at the end of a test. The exchanger reduces the water temperature to less than 250 C in 5 minutes and below 180 C in 15 minutes. Relative corrosion rates of bare uranium at these temperatures compared to 300 C are 25% at 250 C and 2% at 180 C. (4) The method of determining when the failure first starts to occur has not been well defined and in these tests periodic discharging of the fuel elements and visible inspection is used. In some of the tests, failure is indicated by a pressure drop increase across the test section or liquid level change in the loop caused by hydrogen from the reaction being separated from the water in a gas trap. These measurements give indication only after considerable reaction has taken place. Primary water recirculation in the loop is accomplished with a Chempump. This pump has given no trouble, and inspection of the usually troublesome bearings after performing about 30 tests reveals little or no wear. The absence of bearing wear, which previously had been the prime operating difficulty, is attributed to a filter used on the bearing and motor cooling line. Standard pump construction includes a small tubing line which connects from the pump discharge, through a heat exchanger, and finally into the rear of the pump unit. The cooled water

(4) Howe, J.P. and Jones, T.O., Coatings and Corrosion, NNES-IV-6A, [REDACTED]

[REDACTED]

flows past the bearings, rotor, and stator before entering the pump suction where it is mixed with the flow stream. A filter using pre-coated diatomaceous earth on a porous stainless steel basket effectively prevents uranium oxide from reaching the pump bearings. A schematic flow diagram of the ELMO-4 Loop is shown in the Appendix.

C. Metallurgical Considerations

The corrosion rate of uranium fuel and the behavior of coextruded fuel shapes at failure is a matter of extreme importance to the fuel element designer. Before a satisfactory fuel element can be designed, the various parameters which affect its failure behavior must be evaluated. The testing reported upon here describes the effect of several of the parameters and illustrates dramatically the influence of heat treatments.

It must be presumed that beta heat treatment of the coextruded fuel is required to insure maximum dimensional stability or freedom from growth and bumping during irradiation. Beta heat treatment develops a random grain orientation, and with a proper cooling rate through the uranium $\alpha \rightarrow \beta$ transformation, a desirable grain size, so that satisfactory irradiation performance with respect to growth and bumping can be achieved. Coextruded fuel specimens have been defect-tested in ELMO-4 in five conditions (Table I, Appendix).

During beta heat treatment of the uranium fuel in Zr-2 clad coextruded fuel material, not only is the uranium fuel subjected to metallurgical changes, but the coextruded uranium Zr-2 bond is also subjected to metallurgical change. Interdiffusion of the uranium fuel and the Zr-2 clad at the uranium beta treating temperatures increases the amount of U-Zr-2 alloy in the bond. The manner in which the coextruded fuel material is cooled during uranium beta treatment affects the mechanical properties and corrosion characteristics of the bond. Figure 1 (Appendix) contains microstructures of the coextruded bonds and Table II (Appendix) lists the shear strength of the U-Zr-2 bond obtained in the various conditions.

D. Test Results

"As Extruded" Rods

Test Assembly RM-154

The first coextruded fuel assembly tested (RM-154) consisted of four 0.63" diameter, 0.030" clad rods, each defected in the side wall with a 0.025" hole (#72 drill) that penetrated just into the uranium. The assembly was exposed for 1, 2 and 4 hours total time at a velocity of 8 fps. After the first hour, three of the rods had ruptured in almost identical fashion. A 3/8" diameter raised mound had formed at the defect point, and had torn over about 3/4 of its base circumference. The fourth rod showed no physical change during the first hour. After the second hour, two of the rods had a second raised and torn mound and the other two rods has a single 3/8" diameter raised mound. After four hours exposure, the appearances of the rods were quite distinct from each other. The defects ranged from a single 3/8" diameter torn-mound to a 4" slitted and raised portion on one of the rods. In all of the rods, the defects after forming the initial 3/8" round area proceeded down the longitudinal axis of the rod rather than circumferentially. The fuel assembly after each successive exposure period is shown in Figure 2 and data are

tabulated in Table III and plotted on Figure 12.

Test Assembly RM-159

The next assembly, RM-159, consisted of four 0.63" diameter rods, each defected in the unbonded end cap area with a 0.025" hole. After the first hour, only two of the rods had ruptured. Both had the end cap about 90% separated and the rupture had progressed 1/2" down the rod. The total weight loss for both rods was 36 grams. After the second hour, still only the two previously affected rods were ruptured and both to a greater extent. Bulging and splitting had proceeded about 1-1/2" lengthwise and only about 1/3 of the circumferential area was involved. There was no indication of any rupture in the other two rods. All four rods were then exposed for two more hours. The two unaffected rods this time ruptured about 1-1/2" in length while the other two rods which were previously affected ruptured to 2-1/2" in length. Figure 3 shows the test assembly after the first and fourth hours of exposure; pertinent data are tabulated in Table IV.

Test Assembly VR-160

Fuel assembly VR-160 contained four rods each with a 0.030" wide slit through the clad and just to the uranium. In two of the rods, the slits were 3" long and in the other two rods the slits extended their entire length. After one hour exposure, a wavy deformation of the Zircaloy-2 cladding along the slit was observed. The swelling was very localized on the slit and did not proceed radially on the rods. The full-length defected rods showed considerable swelling at the end caps which had occurred adjacent to the slits, but on the short slits the rupturing had progressed longitudinally from the ends of the slits in the form of mounds. Rupturing in the full-length slit pieces progressed from the end caps toward the center of the rod and not necessarily along the original defect. Figure 4 shows these 4 rods after the 2nd hour of exposure; data are tabulated in Table VI.

7-Rod Cluster Assemblies VR-161, VR-162

Two seven-rod cluster fuel elements of the same dimensions as those used in the KER Loops were also tested (VR-161, 162). Each element had only two coextruded fuel rods, the remaining rods being stainless steel. The fuel rods were 0.593" O.D. and were in the as-extruded condition. In both elements the uranium rods were located opposite each other and each was defected with one 0.025" hole which faced the process tube. In VR-161 the Zircaloy-2 clad thickness was 0.020" and in VR-162 it was 0.030". Spacing between rods was 0.070". The primary purpose of the test was to determine if either of the elements would stick in the 2.08" I.D. Zircaloy-2 process tube after failing. Both elements were removed and examined after exposures of 1, 2, and 3 hours. At no time did either of the elements stick in the tube. The defected rods appeared the same after the first hour, each having a single raised and torn mound about 3/8" diameter. The maximum diameter of each element at the ruptured areas was 2.08". After the second and third hours, one of the rods of VR-161 displayed about 3" of longitudinal rupturing, but the rupture in the other defected rod had not increased in extent. The rods in VR-162 did not change in appearance during the additional exposure but did suffer further weight loss.

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7-Rod Cluster Assembly VP-164

Rods in the as-extruded condition were also tested as a wire-wrapped seven-rod cluster fuel element (VP-164). Two opposing rods were each defected with a single 0.025" hole pointed toward the process tube. Each rod was 0.593" diameter and the spacing between rods was 0.070". During the first hour exposure both rods ruptured, one with a 3/8" diameter raised portion and the other with a ruptured area about 3/4" by 1". The fuel element was stuck in the 2.08" I.D. Zircaloy-2 process tube and required about 700 pounds force to remove it. The sticking was caused by a raised "bridge" in the Zircaloy cladding. This "bridge" was about 1" long, 1/4" wide and attached on both ends. There was a hollow area under the bridge. The bridge was pushed down slightly to permit recharging into the process tube and the element exposed for a second hour. This time the element was easily removed and no further increase in the size of the defects was noted. Further uranium corrosion was evidenced by weight loss, however.

Beta Treated Rods

Test Assembly VP-163

Two beta treated rods were tested for comparison with two as-extruded rods. The fuel assembly, VP-163, consisted of two as-extruded rods and two beta treated and air cooled rods (Condition 3, Table 1, Appendix), defected with slits 3" long, 0.030" wide and 0.30 inches deep. After one hour exposure at 300 C, a definite improvement in behavior was shown by the beta treated rods in which a small amount of swelling occurred only at the ends of the slits. The as-extruded rods had raised and torn mounds at the ends of the slits. After three hours total exposure, the as-extruded rods had ruptured severely with the rupture progressing to the end cap in three of the four ends. The swelling of the beta treated rods did not increase during the additional exposure as may be seen in Figure 5. Fuel weight losses in all four rods, with the severe type of defect, were about the same. However, corrosion in the beta treated, air-cooled rods did not produce significant distortion, while equivalent corrosion in the as-extruded rods was accompanied by a high degree of distortion. Pertinent data are tabulated in Table 4.

Test Assemblies VP-165, VP-166

Two fuel assemblies of 4 rods each, with rods in the beta treated, air-cooled condition, were tested with defects similar to those of previously mentioned assemblies RM-154, RM-159, and VP-160 (all as-extruded). Assembly VP-165 contained 2 rods defected with a 0.025" hole in the side wall and 2 rods defected with a 0.025" hole in the unbonded end cap closure. One hour exposure produced a single raised 3/8" diameter mound on each of the side defected rods. Two hours produced no additional change in one of the rods, but the other rod had a series of four raised mounds running longitudinally down the rod. Three hours exposure produced no further effect upon the rod with the single mound, but the other rod had added two more mounds, making a total of six. Four hours exposure produced no additional superficial damage to the rod with one mound, while raising several more small mounds on the other rod. The top assembly of Figure 6 shows the two side-defected rods after two hours exposure. The end-defected rods of this assembly showed no sign of rupture after one hour exposure, and after two hours only one end cap had separated, and this with a minimum amount of damage. Two additional hours (4 total) caused the end cap of the second rod to separate, also with a minimum amount of damage. Data are tabulated in

Tables III and IV, and plotted on Figure 14.

Assembly VR-166 contained two rods defected with 3" long, 0.030" wide slits to the uranium and two rods with full length 0.030" wide slits to the uranium. Of the two rods defected with the three inch slits, only one of the rods developed any serious swelling, and that was at the end of the slit, after one hour. After two hours, both rods had one raised and torn mound at one end of the slit. The other end of the slits had only minor swelling. One unusual aspect of these rods was the absence of swelling along the slits even though a large amount of uranium was exposed. The full length slitted rods had separated end caps after the first hour and a slight amount of swelling extending back from the end caps after the second hour. The maximum diameter of the full slitted rods was less than the 3" slitted rods because no large bulge occurred at the slit ends. VR-166 is shown as the bottom 4 rods of Figure 5 and this should be compared with Figure 4. Data are tabulated in Table IV.

Test Assembly VR-168

The effect of variations in beta treatment schedule on the defect behavior of coextruded rods was also tested. Rods were beta treated and water quenched (Condition 2, Table 1, Appendix) and beta treated, quenched and held at 600 C, and air cooled (Condition 4, Table 1, Appendix). Test assembly VR-168 consisted of 4 rods, two in each of the above heat treated condition, defected with 0.025" side wall defects. After one and two hours exposure, three of the rods, two isothermal treated and one water-quenched, appeared almost identical. A 3/8" diameter mound formed during the first hour and did not increase in size during the second hour. The fourth rod, beta heat treated and water quenched, displayed a failed 3/8" diameter mound and an intact 3/8" diameter mound after one hour. During the second hour severe rupturing occurred with radially swelling extending two inches. The maximum diameter of the rod was 0.90". Failure occurred in one direction from the original defect. The top 4 rods in Figure 3 show the described condition of this test assembly and data is tabulated in Table III and plotted on Figure 12. An additional hour exposure (3 hr. total) did not superficially affect the 3 slightly damaged rods, but extended the failure in the fourth rod to such a degree as to break off the end cap.

Test Assembly VR-169

Test assembly VR-169 consisted of 4 rods, two in each of the above heat treated conditions (see VR-168), defected with 3" long, 0.030" wide, 0.30" deep slits. After one and two hours exposure, three rods, two isothermally treated and one water quenched, appeared almost identical. Only slight damage occurred at the end of the slits, the clad having raised about 0.05" in a 1/8" diameter area. In spite of the small damage, a large volume of fuel had been corroded from the rods. The fourth rod, beta treated and water quenched, suffered severe swelling and splitting of the Zircaloy-2 jacket after one hour. At both ends of the slit, failure progressed radially around the entire rod. Failure at one of the ends had progressed about 1-1/2" in length and had caused the end cap to become separated. At the other end, the rupturing was about 1" in length. After two hours the quantity of swelling had increased and only about 1-1/2" out of the 6" length was not swollen and torn. This rod was in the worst shape of any

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that had been tested. An additional hour of exposure (3 total) did not superficially affect the 3 slightly damaged rods and they appeared as in Figure 4 (bottom 4 rods - VP-169). Data are tabulated in Table 4.

"As Extruded" Tubes

Test Pieces RM-157, RM-158

Coextruded tubular specimens 1.785 O.D., 1.380 I.D., 6" long with 0.030" clad thickness were tested in the as-extruded condition. RM-157 was defected through the O.D. side wall with a 0.025" hole to the uranium. It was exposed for 2 hours in 300 C water, with a water velocity of 11 fps. The ruptured area of the element appeared as a 2-1/2" diameter circle centered on the original defect. This area was comprised on many small individual mounds which had raised and torn. The element did not stick in the 2.1" I.D. Zircaloy-2 process tube after the exposure. One interesting observation was that no swelling had occurred on the inside surface of the tubular specimen. The appearance of RM-157 is shown in Figure 8.

RM-158 was defected in the juncture between the Zircaloy-2 end cap and the uranium tube with a 0.025" hole. It also was exposed for two hours in 300 C water. Upon removal, it was seen that both ends of the tube had ruptured. Metallographic examination revealed a badly contaminated weld which failed by corrosion at the undefected end cap. On each end, swelling occurred over the entire circumference of the tube and extended 2-1/2" on one end and 1-1/2" on the other end. The specimen had washed out of the 2.1" I.D. Zircaloy-2 tube during its exposure and its swelling was unrestrained. The maximum diameter was 2.44". However, the oxide had been washed from under the ruptured portion and the Zircaloy-2 cladding could easily have been pushed into a smaller diameter. The appearance of RM-158 is shown in Figure 8; data are tabulated in Table 4.

Beta Heat Treated Tubes

Test Pieces RM-156 A, RM-156

Coextruded tubular specimens 1.785" O.D., 1.380" I.D., with 0.030" clad thickness were tested in the beta treated, furnace cooled condition (condition 5, Table 1, Appendix). RM-156A was defected through the O.D. side wall with a 0.025" hole to the uranium. After one hour, a single 7/8" diameter raised and torn mound had formed. This mound centered at the hole and raised about .15 to .20 inches above the unaffected surface. There was a cavity beneath the defect. No visible change in appearance after additional exposures of one and two hours was noticed. The weight did decrease during the additional exposure, indicating that fuel was corroding. The appearance of RM-156A after two hours is shown in Figure 9, and it appeared the same after three hours total exposure.

RM-156 was defected at the unbonded end cap with a 0.025" hole. After 2 hours exposure, it was observed that both end caps had separated, but with a minimum of damage. The maximum diameter of the tube was 1.85". Metallographic examination revealed a badly contaminated weld which failed by corrosion at the undefected end cap, such as with RM-158. The appearance of RM-156 after two hours exposure is shown in Figure 9; data are tabulated in Table 4. The elements shown in Figure 9 contrast sharply with those in Figure 8.

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Test Group VP-167

Five tubular specimens 2" long, 1.82" O.D., 1.45" I.D., 0.020" clad were tested as VP-167 in each condition listed in Table 1, Appendix. Each was defected on the O.D. with a 0.025" diameter hole. After the first hour exposure, the isothermal treated tube displayed no failure indications. The other tubes displayed damage ranging from a 1/2" diameter mound to cracks extending to the end caps of the tube. The tubes are listed below in order of best to worst: (1) isothermal treated, (2) air cooled, (3) furnace cooled, (4) as-extruded and (5) water quenched. After two hours, two of the tubes, isothermal treated, and air-cooled, appeared quite similar with each having a 1/2" diameter mound. The other three tubes displayed extensive damage which reached the end caps and as a consequence, most of the surface of the tubes was affected by swelling and tearing of the Zircaloy-2 cladding. The appearance of the five tubular specimens after the first and second hour exposure is shown in Figure 12. Samples 3 and 4 were tested to a total exposure of 4 hours and there was no change in appearance, but they continued to lose fuel by corrosion as revealed by weight loss. Damage to these specimens was confined almost exclusively to the outside surface. Data is tabulated in Table III and plotted on Figure 14. 15

General

Dimensional and weight changes following each exposure are to be found in the Appendix in Tables III - VI and Figures 12 and 13. The effect of heat treatment upon failure behavior is quite dramatic. Water quenched specimens (Condition #2) appear to corrode rapidly with a high degree of clad deformation. However, the anomalous behavior of several water-quenched rods (Figure 8) requires that this type of heat treatment be examined more closely since an equivalent or superior heat treatment to condition #4 may be developed. Isothermally treated rods and tubes (Condition #4) behave in the most desirable manner from the standpoint of fuel corrosion during failure and the degree of accompanying clad distortion.

"Incubation" periods were noted for some types of defects during the tests. Pin-hole type defects through the bonded clad require some period of exposure before the onset of fuel loss and rod damage. Parallel studies in autoclaves have shown that these incubation periods are a function of test temperature and nature of the bond.⁽⁵⁾ The incubation period for pin-hole type defects in the unbonded end cap areas may be purely mechanical in nature (i.e., caused by oxide or internal gas pressure plugging).

Cleavage type failures were simulated in some of the tests. The basic idea was to expose a large amount of the uranium in two opposing sides of a slit with a minimum space between the uranium surfaces. The defect actually used was a machined slit 0.030 inches wide through the cladding to a depth of 0.30 inches in the uranium. With the small diameter rods, the slit depth was half of the rod diameter. The results were quite surprising in that relatively minor damage was done to the rod during the exposure. In all cases, if the damage was severe, it was caused by swelling at the end of the slit and not

(5) Troutner, V.H., Interim Report #2, Rupture Kinetics, HW-61379, August 3, 1959, (Unclassified).

along the length of the slit. In one case, with an isothermally treated rod, there was no swelling after three hours exposure, even though half of the uranium had been removed by corrosion.

E. Uranium Oxide Particle Analysis

Particle size of the uranium oxide formed from a rupture is important in the operation of the NPR. Whether the oxide from ruptures can be easily washed through the piping systems or whether it will collect in low velocity sections will affect the decontamination of the system greatly. Also, rupture detection may depend on the rupture particulate matter going past the monitor. At low water temperatures where the corrosion product release rate may be quite low, settling of particulate matter before reaching the monitor could make the task of determining the rupture location exceedingly difficult because of the small amount passing the monitor at any time. Further, the mixing of water from tubes feeding an effluent header would dilute any rupture activity in the header and make more difficult the task of monitoring.

Uranium oxide samples were taken from four different locations in the loop following a rupture test with coextruded rods at a water temperature of 300 C. Location of the sampling points are described in Table III and illustrated in the schematic diagram of ELMD-4, page 14; the particle size analysis is also shown in Table VII. As can be seen from the data in Table VII, the particles are fine with the bulk being between 5-20 microns. Being so small, the particles do tend to follow the flow stream and unless the water velocity drops severely, they will not settle out. The water velocity at the exit of the ELMD-4 test section after leaving the restricted fuel element and process tube and entering the three inch pipe is estimated at 1.5 - 2.0 fps. Most of the uranium oxide from any of the experiments is scattered throughout the loop and not concentrated in the test section outlet. However, some of the oxide tends to deposit on the pipe walls from which it can be removed, either by swabbing as in the ELMD-4 Loop or by chemical decontamination as in the IRP-1 Loop.

F. Possible Failure Mechanisms

STOP

To simplify NPR fuel heat treatment schedules, and to optimize that heat treatment for the best failure behavior, it is desirable to understand the mechanisms of failure. Several ideas have been developed, based upon published knowledge of the U-Zr system and upon observations made in the defect testing program, regarding the failure mechanism.

The idea has often been expressed that the strength of the clad-to-fuel bond may be of utmost importance in determining failure behavior. Bond strengths (in shear) have been shown to vary, depending upon their heat treatment and resulting structure, from 70,000 psi downward to 13,000 psi (see Table II). The bond strength tends to decrease with increasing bond zone thickness and may vary for a given bond thickness depending upon the metallurgical nature of the bond. Test results indicate that the best performance is not obtained when the bond strength in shear is greatest, but that some intermediate bond strength may be desirable.

(6) Personal communication, G.E. Neibaur.

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The possibility exists that some mechanical condition or structural feature in the bond which alters the bond strength may also alter the bond zone corrosion behavior quite significantly. For example, bonds in the uranium beta-treated, water-quenched condition are extremely strong, but their failure performance varies from satisfactory to poor. The U-Zr alloy at some zone in the bond is probably very hard, brittle, and highly stressed because of partial phase transformations. The hard and brittle bond may promote easy propagation of bond line separation from corrosion product build-up. The highly-stressed condition in the bond may make it susceptible to reaction with hydrogen liberated by uranium corrosion, so that bond line failure occurs rapidly. Metallographic examination of such bonds has not revealed the mechanism of bond failure. Bonds in the as-extruded condition also behave poorly, even though they are of high strength but probably not brittle or highly stressed. A possible explanation for their poor performance may be that a relatively small amount of hydrogen from uranium corrosion is required to react with the extremely thin bond so that bond line failure occurs rapidly.

Bonds which have been uranium beta treated and slow-cooled behave in a much better manner than as-extruded or water-quenched bonds. In the slow-cooled condition, there is less U-Zr alloy high temperature phase retained and transformation to lower temperature phases is more complete, hardness and stresses are lower, and a zirconium-bearing phase has begun to develop, or precipitate, in the uranium side of the bond (Figure 1). Complete transformation with decreased hardness and reduced stresses and a more developed precipitate in the uranium are promoted further by an isothermal treatment at about 600 C with a consistently superior failure behavior being observed. The improved performance may result because the mechanical conditions of the bond have been improved and its susceptibility to hydrogen attack has been decreased, even though the bond is weaker. It is further possible that the zirconium bearing phase which precipitates in the uranium side of the bond may act as a getter for hydrogen liberated by uranium corrosion and thus tend to arrest hydrogen attack at the bond line.

G. Acknowledgement

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OPERATING CONDITIONS

TEMP. 300°C - PRESS. 1500 P.S.I.

FLOW. 50 G.P.M

SCHEMATIC DIAGRAM OF ELMO-4 LOOP

Cross-Section of Coextrusion Bonds

Remarks

Zr-2 Clad Bond U Fuel	"As extruded" condition. The bond is no more than a line of demarcation between the fuel and the clad. High bond strength is obtained, but high corrosion rate with severe clad deformation results. <u>250X</u>
Zr-2 Bond U	Beta treated water quenched. The bond has thickened from virtually zero to about 0.00025 inch. A high bond strength is obtained, but high corrosion rate with severe clad deformation results. <u>250X</u>
Zr-2 Bond U	Beta treated, furnace cooled (vacuum). Air cooled and isothermally treated bonds appear similar to this. A lower, but respectable bond strength is obtained. Lower corrosion rates with a minimum of clad deformation results. Bond about 0.00015 inch thick. <u>1000X</u>
Zr-2 Bond U	Diffusion treated to produce thick bond about 0.008 inch and oil quenched. Low bond strength obtained. Bond fracture propagates rapidly permitting high corrosion rate and severe clad deformation. Specimens in this condition have been tested in autoclave, but not in ELMO #4. <u>100X</u>

FIGURE 1

Photomicrographs of Coextruded Bonds in Various Conditions

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FIGURE 2

RM-154 Coextruded Rods in the As-extruded Condition After one, Two, and Four Hours Exposure to 300C Water. Initial Defect in Each Rod was a 0.025 Inch Pinhole.

FIGURE 3

RM-159 Coextruded Rods in the As-extruded Condition After One and Four Hours Exposure. Initial Defect was a 0.025 Inch Pinhole in the End Cap Void.

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FIGURE 4

VP-160 Coextruded Rods in the As-extruded Conditions After Two Hours Exposure.
Initial Defect was a 0.030 Inch Wide Slit Down to the Uranium.

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FIGURE 5

VP-163 Coextruded Rods After Three Hours Exposure. Initial Defect was a 0.030 Inch Slit, 0.3 Inches Deep. Top Two Rods are Beta Heat Treated and Air Cooled, Bottom Two Rods are in the As-extruded Condition.

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FIGURE 6

VP-165 Top and VP-166 Bottom, Coextruded Rods Beta Heat Treated and Air Cooled After Two Hours Exposure. Initial Defects were 0.025 Inch Pinholes, Top, and 0.030 Inch Wide Shallow Slits, Bottom.

FIGURE 7

VP-168 Top and VP-169 Bottom Coextruded Rods After Two Hours Exposure. Top Two Rods in Each Element were Beta Heat Treated, Isothermally Treated and Air Cooled. Bottom Two Rods were Beta Heat Treated and Water Quenched. VP-169 Slits were 0.30 " Deep.

FIGURE 8

RM-157 Top and RM-158 Bottom, Coextruded Tubes in the As-extruded Condition After Two Hours Exposure. Initial Defect was a 0.025 Inch Pinhole.

FIGURE 9

RM-156 and VP-156A. Coextruded Tubes, Beta Heat Treated and Furnace Cooled, After Two Hours Exposure. Top Element Defected in the End, Bottom Element Defected in the Side with 0.025 Inch Pinholes.

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FIGURE 10

VP-167 Coextruded Tubes After One and Two Hours Exposure. Each was Defected with a 0.025 Inch Pinhole. From Left to Right: (1) As-extruded, (2) Beta Heat Treated, Water Quenched, (3) Beta Heat Treated, Air Cooled, (4) Beta Heat Treated, Isothermally Treated and Air Cooled, (5) Beta Heat Treated, Furnace Cooled.

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- as extruded + cool. 1 (Table 1)
- + 6 - H₂O quench - cond. 2
- △ 6 - air cool - cond. 3
- × 6 - isothermal treat - cond. 4
- 6 - furnace cool - cond. 5

VR-167
1 tube each
condition

FIGURE 11 Weight loss of coextruded Uranium-Zr-2 tube specimens (VR-167), 1.82" O.D. x 1.45" I.D. x 0.020" clad defected with 0.025" dia. hole in side wall, at 300°C, 1500 psi, in ELMO-4.

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FIGURE 14

Weight loss of coextruded Uranium-Zr-2 rod specimens, 0.63" dia. x 0.030" clad, defected with 0.025" dia. hole in side wall, at 300°C, 1500 psi, in ELMO-4.

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TABLE I

Conditions of U-Zr-2 Coextruded Fuel Specimens Defect Tested
in ELMO-4 High-temperature, High-pressure Loop

<u>Condition</u>	<u>Description</u>
1. As extruded	As fabricated fuel, coextruded in the high α uranium range (640 C).
2. β treated, water quenched	Heated for 10 minutes in salt bath at 720-730 C (beta uranium), quenched in cold water.
3. β treated, air cooled	Heated for 10 minutes in salt at 720-730 C, cooled to near room temperature in air.
4. β treated, isothermal treatment, air cooled	Heated for 10 minutes in salt at 720-730 C, quenched in salt at 590 - 600 C and held for 10 minutes, air cooled to near room temperature.
5. β treated, furnace cooled.	Heated for a nominal (*) 10 minutes in a vacuum furnace at 720 - 730 C, furnace cooled to near room temperature.

(*) described as nominal since the control of time at temperature was poor.

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TABLE ~~II~~ VII

Coextruded Bond Shear Strength Values
At Room Temperature

Condition of Sample	Number of Samples	Average Shear Strength psi	Remarks
As extruded	6	Ave. of 3 69,250	These three failed outside the bond by shear in the Zr-2 clad and in the U.
		Ave. of 3 55,000	These three failed by tension in the Zr-2 clad.
Salt bath β treated, 10 m. @ 720-30 C, H ₂ O quench	2	Ave. of 2 71,000	Failed by shear in Zr-2 clad.
Salt bath β treated, 10 m. @ 720-30 C, air cooled	2	Ave. of 2 61,050	Failed by shear in Zr-2 clad with some tendency to fail in bond.
Salt bath β treated, 10 m. @ 720-30 C, quenched to 600 C, held for 10 m., air cooled	3	Ave. of 3 57,775	Failed partially by shear in Zr-2 clad with some tendency to fail in bond.
Vac. β Treated, 10 min. @ 720-30 C, furnace cooled	2	Ave. of 2 47,750	Failed predominately in the bond.
Vac. Diff. HT, 62 hrs. @ 840 C, furnace cooled	1	27,800	Failed in bond adjacent to U.
Vac. Diff. HT, 62 hrs. @ 840 C, furnace cooled, 2 hrs. at 840 C, oil quenched	1	12,870	Failed in bond adjacent to U.

Section view of bond shear strength specimen -

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TABLE III. 1

Diameters of Coextruded U-Zr-2 Rod and Tube
Specimens Defected With 0.025" Diameter Hole
In Can Wall After Exposure In ELMO-4 at 300 C

Rods - 0.63" dia. x 0.030" Clad

Test No.	Condition (See Table I)	Dia. After Exposure, Inches			
		1st Hr.	2nd Hr.	3rd Hr.	Max.
RM-154 ¹	as extruded - 1	0.85	0.86	0.87(4 hr)	0.94
VR-168 ²	B - H ₂ O quench-2	0.76	0.83	0.88	0.90
VR-165 ²	B - air cooled-3	0.83	0.83	0.83(4 hr)	0.86
VR-168 ²	B - isothermal treat-4	0.75	0.76	0.76	0.76

1-4 rods, average data

2-2 rods, average data

Tubes - 1.82" O.D. x 1.45" I.D. x 0.020" Clad

Test No.	Condition (See Table I)	O.D. After Exposure, Inches			
		1st Hr.	2nd Hr.	3rd Hr.	Max.
VR-167 ¹	as extruded - 1	1.97	1.97	2.07	2.07
VR-167 ²	B - H ₂ O Quench-2	1.98	2.1	2.1	2.1
VR-167 ²	B - air cool - 3	1.9	1.9	1.9	1.9
VR-167 ²	B - isothermal-4	1.82	1.95	1.95	1.95
VR-167 ³	B - furnace cool-5	1.96	2.00	2.00	2.08

1 - 1 tube, RM-157 also tested in this manner.

2 - 1 tube

3 - 1 tube, RM-156A also tested in this manner.

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TABLE IV II

Diameters and Weight Losses of Coextruded
U-Zr-2 Rod and Tube Specimens Defected With
0.025" Diameter Hole In Unbonded End Cap Area
After Exposure In ELMO-4 At 300 C.

Rods - 0.63" O.D. x 0.030" Clad

Test No.	Condition (See Table I)	Dia. Inches				Wt. loss - gram		
		1 hr.	2 hr.	4 hr.	Max.	1 hr.	2 hr.	4 hr.
RM-159 ¹	As extruded-1	.76	.77	.80	.82	18	52	100
VP-165 ²	B - air cool-3	-	.71	-	.71	-	0	36

1-4 rods, average data

2-2 rods, average data

Tubes - 1.785" O.D. x 1.380" I.D. x 0.030" Clad

Test No.	Condition (See Table I)	Dia. Inches			Wt. loss - grams	
		1 hr.	2 hr.	Max.	1 hr.	2 hr.
RM-158 ¹	As extruded-1	-	2.44	2.44	-	268
RM-156 ¹	B - furnace cooled	-	1.84	1.85	-	57

1 - 1 tube

TABLE IV III

Diameters and Weight Losses of Coextruded U-Zr-2
0.63" Diameter Rods Defected With a 3" Long, 0.030"
Wide, 0.30" Deep Slit After Exposure In ELMO-4 At 300 C

Test No.	Condition (See Table I)	Dia. Inches				Wt. loss - grams		
		1st hr.	2nd hr.	3rd hr.	Max.	1st hr.	2nd hr.	3rd hr.
VP-163 ¹	As extruded-1	.75	.81	.82	.83	93	246	335
VP-169 ¹	B-H ₂ O Quench-2	.77	.78	-	.90	98	270	-
VP-163 ¹	B-air cool-3	.68	.68	.69	.71	98	231	288
VP-169 ¹	B-isothermal - 4	.66	.66	.66	.67	73	175	222

1 - 2 rods, average data

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TABLE VI IV

Diameters and Weight Losses of Coextruded U-Zr-2
0.63" Diameter Rods Defected With 0.030" Wide Slits
Through The Clad To The Uranium After Exposure In
ELMO-4 at 300 C.

Rods With 3" Length Slits

Test No.	Condition (See Table I)	Dia. - Inches			Wt. loss-gram	
		1 hr.	2 hr.	Max.	1 hr.	2 hr.
VP-160 ¹	As extruded - 1	—	.72	.73	—	—
VP-170 ²	B-H ₂ O quench-2	.68	.71	.77	36	136
VP-166 ¹	B-air cool-3	.73	.80	.81	39	185
VP-170 ²	B-isothermal-4	.67	.67	.68	26	94

1 - 2 rods, average data

2 - 2 rods, average data, VP-170 not described in text

Rods With Full Length Slits

Test No.	Condition (See Table I)	Dia. Inches			Wt. loss-grams	
		1 hr.	2 hr.	Max.	1 hr.	2 hr.
VP-160 ¹	As extruded - 1	—	.77	.83	—	—
VP-166 ¹	B - air cool-3	.68	.70	.71	91	198

1 - 2 rods, average data

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TABLE ~~VII~~ VI

Particle Size Analysis of Four Uranium Oxide Corrosion
Product Samples Taken From ELMO 4 After a Coextruded
Fuel Rod Rupture Test At 300 C, 1500 psi.

Sample 1. Outlet end of the test section (low velocity).

Sample 2. Inside the 100 mesh strainer (before passing through)

Sample 3. Main loop drain.

Sample 4. Chempump cooling water line filter.

} See Schematic of
ELMO-4 for location.
Figure 1,

<u>Particle Size</u> <u>Range (Microns)</u>	<u>Percent of Sample Within Range</u>			
	<u>Sample 1</u>	<u>Sample 2</u>	<u>Sample 3</u>	<u>Sample 4</u>
1.5-5	11	3	0.6	3
5-10	34	20	6	7
10-15	26	18	52	40
15-20	18	19	29	28
20-25	7	6	7	8
25-30	2	2	3	2
30-40	2	2	1	
40-50		0.4		
> 30				2
> 40	0.2		0.7	
> 50		31		
Total	<u>100.2</u>	<u>101.4</u>	<u>99.3</u>	<u>90</u>

DATE

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8/24/94

END