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EMBRITTLEMENT OF ZIRCALLOY-CLAD FUEL RODS
BY STEAM DURING LOCA TRANSIENTS

D. O. Hobson
P. L. Rittenhouse

OAK RIDGE NATIONAL LABORATORY
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EMBRITTLEMENT OF ZIRCALOY-CLAD FUEL RODS BY STEAM
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ABSTRACT

One of the possible consequences of a loss-of-coolant accident (LOCA) in a light-water reactor would be the rapid heatup of the fuel tubes in a steam environment. During the period of time in which the cladding was exposed to steam (provided both from boiloff of residual water in the pressure vessel and by the emergency coolant) at elevated temperatures, reaction of the Zircaloy with the steam would cause the formation of an oxide film on the cladding surface and an oxygen gradient into the wall. Since the ductility of Zircaloy is a direct function of oxygen content, and since the oxygen content is a function of time and temperature of steam exposure, a study of the degree of embrittlement to be expected in a LOCA necessitates the examination of specimens exposed to specific temperatures for given lengths of time in a steam environment. Correlations are developed among steam exposure, oxygen content, ductility, and hardness which permit estimating the degree of embrittlement the cladding would attain during any LOCA transient. Curve fitting to LOCA transient data and subsequent integration provide easy analysis of the amount of embrittlement expected.

INTRODUCTION

The design-basis accident for a light-water reactor is the double-ended rupture of one of the main inlet coolant-water pipes to the primary system pressure vessel, with resultant blowdown of the coolant inventory and initiation of fuel rod heatup. While the probability of the occurrence of an accident of this severity is extremely small, the potential consequences are so severe it must be considered. This accident defines the performance requirements of many of the engineered safety features provided to mitigate the consequences of all credible accidents. We use as the starting point for our study the assumption that such an accident has occurred. Assumptions are also made concerning the efficacies of the emergency core cooling systems (ECCS) to provide

the basis for selection of various heatup rates and peak turnaround temperatures. Our main concern will be the amount of embrittlement the fuel cladding could sustain during specific loss-of-coolant accident (LOCA) transients. Embrittlement is of concern because of the need to maintain core integrity during and after a LOCA. Local embrittlement could cause fragmentation of some of the fuel rods with the fragments then causing additional blockage of the coolant channels. Gross embrittlement of the core could cause core slumping and complete loss of core geometry.

The range of temperatures considered in this study is 1600 to 2500°F. The 1600°F temperature represents approximately the minimum temperature for metal-water reaction. The 2500°F temperature represented the highest temperature at which we could obtain accurate short-time exposures. This temperature range includes the maximum transient temperatures calculated, in the literature, for LOCA's.

The reaction of steam with the Zircaloy cladding is one of dissociation of the steam into oxygen and hydrogen. The oxygen reacts with zirconium according to $\text{Zr} + \text{O}_2 \rightarrow \text{ZrO}_2 + \text{heat}$ [$> 10^6$ Btu for 1% metal-water reaction in 1000-MW(e) reactors].

To illustrate the various events postulated for LOCA transients, we show Fig. 1. In order to maintain generality, the temperature and time axes have deliberately been left unscaled; the curves generalize conditions for either a pressurized-water reactor (PWR) or boiling-water reactor (BWR) LOCA. The sequence of overlapping time-dependent events is blowdown, cladding heatup by redistribution of fuel rod energy, heatup by fission-product decay, and exothermic metal-water reaction. As the fuel tube heats up as a function of time, there are several temperature dependent changes that can take place in the cladding: cladding deformation and rupture over a wide, pressure-dependent range; eutectic formation with non-Zircaloy core components such as Inconel grid spacers; and, at elevated temperatures, significant metal-water reaction. If not terminated, the transient would eventually reach temperatures high enough to melt any metal remaining in the cladding. The heatup of the cladding in Fig. 1 is shown as being turned around by each of three arbitrary transients indicated by dashed lines. The lower, "ideal,"

Fig. 1. Generalized LOCA Transient Showing Sources of Heat and Some of the Temperature-Dependent Consequences of Core Heatup.

transient is one in which the LOCA is terminated before cladding rupture occurs; fission products are still contained in the fuel rods. Although very desirable from the standpoints of fission-product containment and coolant channel blockage, it is considered unreasonable to expect such a low turnaround in a major LOCA. Current ECCS design calls for transient turnaround as shown by the middle dashed curve, which allows rod rupture but very little metal-water reaction. The upper dashed curve is for a transient in which large amounts of metal-water reaction occur, resulting in considerable embrittlement.

EXPERIMENTAL PROCEDURE

This study will define the time-temperature relationships between steam exposure and embrittlement. The steam-exposed tubes were evaluated on the basis of hardness and oxygen penetration as functions of times at temperatures. There are several ways to evaluate such data, but the methods of the most practical value for relating the results to hypothesized accident situations are those that can show mechanical properties as functions of specimen history. In this study, we have sought a correlation between exposure times and temperatures and both microstructural and mechanical changes in the material. We chose to evaluate embrittlement due to oxidation of both surfaces of the tube. Such an evaluation is conservative relative to unruptured tubing (which is unoxidized on the inner surface) in a LOCA and nonconservative relative to ruptured, thinned down tubing.

Nineteen steam-oxidized and water-quenched PWR-size, reactor-grade Zircaloy tubes (0.422 in. diam $\times$ 0.027 in. wide) were used as the basis for our embrittlement evaluation. These had been exposed to a steam environment at nominal temperatures from 1700 to 2500°F for times ranging from 2 to 60 min, depending upon temperature. A Globar-type furnace was used to heat a ceramic muffle tube inside which the specimen was suspended. The furnace was calibrated so that the temperature at the mid-length of the specimen could be monitored indirectly. The tube was held at temperature in steam for a specified length of time after which it was dropped, through an opened ball valve, into water. The steam supply was such that the reaction was not steam-limited. The furnace characteristics caused a decreasing temperature gradient to exist from the center of the tube length toward each end. However, furnace temperature scans showed the gradient to be less than 10°F over a 3-in. tube length at the center line. In order to obtain a correlation between exposure temperature and tube properties, a ring cut from the midpoint of each 18-in.-long tube was mounted, polished, photographed in polarized light (unetched), and hardness tested. Data from these rings were considered as having been produced at the reported temperatures and times in this report. Eight other rings, each 1/4 in. long, were cut from near the

center portion of each tube. A testing device was designed and built capable of producing compressive diametral strains in these ring specimens to an accuracy of ± 0.001 in. of compressive deflection. This device was mounted inside a furnace capable of a maximum temperature of 1900°F . Figure 2 illustrates the testing device. The deflections were produced under impact loading to simulate stress conditions that could exist due to steam bursts, hydrodynamic forces, or tube ruptures during the LOCA.

Changes in the microstructure of the tube walls were evaluated metallographically to determine the extent of oxidation, and these changes were correlated with exposure times and temperatures. The relationships derived from this study, although based on isothermal exposures, were directly amenable to integration over any given LOCA transient to provide estimations of the amount of embrittlement caused by that transient.

RESULTS

Metallography and Hardness Results

The effects of steam exposure were evaluated metallographically to determine the rates of migration of the brittle phases into the ductile wall material. This information is the foundation for our evaluation and also allows comparison with data reported by others. Microhardness measurements were taken to establish a correlation between hardness level and embrittlement and to provide an estimation of the oxygen distribution through the tube wall.

Figure 3 is an idealized cross section through the wall of a steam-oxidized tube, while Fig. 4 is a micrograph of the cross section of a tube exposed to steam for 4 min at 2400°F . In this study, oxygen was diffused into both surfaces of the tubes, creating outer layers of ZrO_2 and subjacent layers of oxygen-stabilized α -zirconium. The thickness of the combined ZrO_2 and α -phase zirconium layers depends on a combination of exposure time and temperature. At sufficiently high temperatures (approximately 1800°F and above) the β -phase exists between the encroaching α -phases. The β -phase transforms back to α upon quenching, but the

Fig. 2. Schematic of Device Used to Test Ductility of Oxidized Ring Specimens Under Impact Loading at Various Selected Temperatures.

Fig. 3. Idealized Cross Section Through the Wall of a Steam-Oxidized Tube Showing the Phases that Will Form. The midwall phase is β at elevated temperatures and transforms back to α -phase on cooling.

Fig. 4. Photomicrograph, Taken With Polarized Light, of a Specimen After Exposure to Steam for 4 min at 2400°F. The various phases are visible, with the oxide seemingly divided by interfaces on each side of the tube wall.

microstructure is quite different from that of the oxygen-stabilized phase. The thickness (ξ) of the ZrO_2/α -phase outer layer measured on our specimens is plotted against the square root of time in Fig. 5. The points form a set of six straight lines whose slope are functions of the test temperatures. Figure 6 is a plot of ξ versus the square root of the product of the diffusion coefficient (D) and time, where D is given by¹

$$D = 1.196 \exp(-41,000/RT) . \quad (1)$$

¹A. W. Lemmon, Jr., Studies Relating to the Reaction Between Zirconium and Water at High Temperatures, Battelle Memorial Institute USAEC Report BMI-1154, pp. 4-8 (Jan. 3, 1957).

Fig. 5. Thickness (ξ) of the Combined ZrO_2/α -Phase Layer Plotted Against the Square Root of Exposure Time. The straight lines are characterized by exposure temperature.

Fig. 6. Thickness (ξ) Plotted Against the Square Root of the Quantity: Diffusion Coefficient times Exposure Time. The equation for the least squares fit of the ORNL data is shown together with the equation for the Idaho line [ref. R. H. Meservey and R. Herzel, Brittle Behavior of Zircaloy in an Emergency Core Cooling Environment, Idaho Nuclear Corporation USAEC Report IN-1389, p. 15 (September 1970)].

In this expression, R is $1.987 \text{ cal/mole}\cdot^\circ\text{K}$ and T is a sample temperature in $^\circ\text{K}$. The points fall in Fig. 5, with some scatter, into a straight line that is nearly identical to the line obtained for specimens of Zircaloy tested under similar conditions by Idaho Nuclear Corporation.² Appendixes A and B contain discussions of oxygen diffusion and α -phase migration as related to previous work¹ and discussion of metal-water reaction calculations.

Figure 7 is a plot of hardness measurements, taken in the middle of the tube walls, as a function of ξ . The midwall hardnesses represent

²R. H. Meservey and R. Herzel, Brittle Behavior of Zircaloy in an Emergency Core Cooling Environment, Idaho Nuclear Corporation USAEC Report IN-1389, p. 15 (September 1970).

Fig. 7. Room-Temperature, Mid- β -Phase Hardness Plotted as a Function of ξ . The least squares equation is given.

the lowest hardness region in the wall, since the oxygen content is lowest there. It is seen that a linear, temperature- and time-dependent relationship is obtained. It will be shown later that specimen ductility is directly related to midwall hardness.

Evaluation of Embrittlement Due to Isothermal Exposure

In any LOCA temperature transient the fuel tubes may become embrittled and, therefore, it is of great importance to determine the relationship between ductility and steam exposure. The ductility of the steam-oxidized tubing was evaluated using the compression apparatus described in "Experimental Procedure," p. 4. Rings of the oxidized tubing were diametrically compressed various amounts at temperatures up to 1900°F. It has been reported² that tubing specimens with ξ values up to approximately 0.035 cm, compared with the maximum ξ of 0.028 cm in this study, were ductile during axial compression testing at 1800°F. However, it must be remembered that, unlike ORNL tubes, these tubes were reacted only on the outer surface.

It is recognized that a section of fuel cladding would not be uniformly oxidized on both surfaces during a LOCA. Depending upon the internal pressure in the rod, rupture would occur over a range of temperatures. Oxidation of the inner surface would not begin until rupture and then would likely be confined to regions extending a short distance on each side of the rupture. However, as the only ductile material remaining in the tube wall is the transformed β -phase, any combination of ξ values subtracts from this β thickness, whether ξ has a value greater than zero only on the outer surface or whether it is some combination of ξ values on both surfaces. In the present study we chose to oxidize uniformly on both inner and outer surfaces. Such a procedure yields conservative results when compared with tubing oxidized, with only the outer surface exposed, to identical conditions of time and temperature. Our results will be shown to be nonconservative in the region of the rupture, where two-surface oxidation combined with wall thinning can be obtained.

Figure 8 is a plot of specimen ductility as a function of deformation temperature and the fraction of wall thickness of the oxidized tubes consisting of transformed β . This fraction (F_W) is defined as the width of the transformed β -phase divided by the wall thickness. The plot is divided into three regions. The right-hand region, characterized by open circles, represents specimens that received relatively low amounts of oxidation. These rings were flattened, diametrically and under impact loading, 0.150 in. with no cracking of the transformed β structure. The ZrO_2 /oxygen-stabilized α layers were cracked, but these represented only a small fraction of the wall thickness. The center region represents rings that exhibited from one to four fractures through the wall, but which also showed some amounts of plastic strain. The left-handed region of the plot represents specimens that showed no plastic deformation whatsoever; after the test, the fragments could be placed together to form an undistorted ring of pretest dimensions. The dashed line separating the zero-ductility region from the partial-ductility region is important because it is independent of the amount of deformation one might wish to impose on the specimen. It, therefore, defines the zero-ductility temperature (ZDT) of the cladding as a function of microstructure and deformation temperature. The dashed curve separating the partial-ductility region from the ductile region is deformation dependent. Its position is determined by the amount of deformation required of the ring specimen.

A further correlation of ductility and deformation temperature was obtained with room-temperature hardness measurements made in the middle of the wall thicknesses of the tested specimens. Figure 9 illustrates the results obtained. The points are as previously defined and the closed, underlined points represent specimens with zero ductility at their indicated deformation temperatures. Each data point represents the average of from four to six hardnesses taken per specimen. The correlation is seen to be quite good.

Figure 8 was used in conjunction with Fig. 5 to develop relationships among exposure time and temperature and deformation temperature from which cladding ductility could be obtained. These relationships are shown in Figs. 10 and 11, which are charts from which the times,

Fig. 8. Specimen Ductility as a Function of Deformation Temperature and Fraction of Wall Thickness (F_W) Consisting of Transformed β -Phase.

Fig. 9. Deformation Temperature Versus Room-Temperature Midwall Hardness. The points have the same nomenclature as in Fig. 8.

Fig. 10. Exposure Temperature Versus Exposure Time, In Steam, With the Curves Drawn to Show Temperatures Above Which the Ring Specimens Were Ductile Under Diametral Impact Strains of 0.150 in.

Fig. 11. Figure Similar to Fig. 10, But With Curves Drawn to Show Temperature Below Which the Ring Specimens Had No Ductility Under Impact Loading.

for a given temperature, to yield "total" ductility and zero ductility, respectively, may be read. As an example, consider a tube isothermally exposed at 2400°F to steam for 5 min. From Fig. 10, one finds that the tube would have total ductility (on being diametrically compressed 0.150 in.) above approximately 1900 to 2000°F. From Fig. 11, one finds that the tube would have zero ductility below approximately 900°F.

Technique for Estimating Embrittlement Caused by Any LOCA Transient

The charts shown in Figs. 10 and 11 are useful for evaluating the effects of isothermal exposures and can also be used to estimate the embrittlement that would occur in a LOCA. We have, however, developed another method for estimating this embrittlement that involves the integration of ξ as a function of temperature over a portion of the time period of the LOCA transient. The slopes of the straight lines in Fig. 5, the plot of ξ versus the square root of time, were plotted as a function of their corresponding temperatures as shown in Fig. 12. The equation for this curve is

$$\left(\frac{d\xi}{d\sqrt{t}} \right)_{\text{isothermal}} = 1.08 \times 10^{-6} T + 5.15 \times 10^{-6} e^{0.0168(T - 2200)} - 1.70 \times 10^{-3} . \quad (2)$$

Previously, we have reported values of ξ calculated by substituting curve-fitted equations for specific LOCA transients for temperature (T) in Eq. (2) and then integrating the resultant equation numerically.³ Such a method, does not take into account prior ξ penetration resulting from other than isothermal exposure. In other words the penetration rate, $\frac{d\xi}{dt}$ or $\frac{d\xi}{d\sqrt{t}}$ may vary appreciably from the isothermal rate, depending upon preexisting values of ξ . We have developed a revised method as described below.

A diffusion-controlled rate of migration of ξ is assumed so that the parabolic equation

$$\xi = \delta t^{1/2} \quad (3)$$

³D. O. Hobson, "Analyses of LOCA Transients in Terms of Cladding Embrittlement," Trans. Am. Nucl. Soc. 14, 700-701 (1971).

Fig. 12. Rate of Change of ξ with Time Versus Exposure Temperature. The curve is linear to 2200°F and then shows a rapid increase. The data are fitted by the equation shown.

is applicable. The coefficient, δ , represents empirical Eq. (2). Equation (3) is applicable, in this form, only under isothermal conditions where time is measured from time zero, that is, from initial clean surface conditions. Equation (3) is differentiated to yield an equation for rate of ξ migration as a function of time, still for isothermal conditions.

$$\frac{d\xi}{dt} = 1/2 \delta t^{-1/2} \quad (4)$$

Solving Eq. (3) for $t^{1/2}$, we substitute its reciprocal, $t^{-1/2}$, into Eq. (4) to obtain

$$\frac{d\xi}{dt} = 1/2 \frac{\delta^2}{\xi} \quad (5)$$

This equation is now applicable to nonisothermal conditions. It incorporates, in its denominator, a ξ term that takes into account preexisting oxidation, so that integration of Eq. (5) is now valid over any portion of a LOCA transient curve.

$$\xi = \int_{t_i}^{t_f} \frac{\delta^2}{2\xi} dt \quad (6)$$

or

$$\xi = \sum_{t_i}^{t_f} \frac{\delta^2}{2\xi} \Delta t \quad (7)$$

We used graphical integration and the equation

$$\xi_{it1} = \xi_i + \frac{\delta^2}{2\xi_i} \Delta t \quad (8)$$

to obtain our results. These, obtained by the above method, produce ξ values larger than those previously reported.³ A simplistic explanation for this can be given. In an isothermal situation the rate of growth of ξ is dependent upon the continuously-forming barrier of oxide and α -phase at that temperature. This rate is continually slowed by the increasing barrier thickness. If the barrier were either thicker or thinner at a given time and temperature than the thickness appropriate to that temperature, the rate would not equal the isothermal rate. Rather, a thinner barrier would produce a faster rate, and a thicker would produce a slower rate of growth of ξ . So, at any point on the increasing-temperature slope of a transient, the ξ value for the previously formed layer is less than for the equivalent isothermally-produced layer for the same exposure time. Therefore, the instantaneous rate $\left(\frac{d\xi}{dt}\right)_{\text{transient}}$ is

greater than $\left(\frac{d\xi}{dt}\right)_{\text{isothermal}}$ and greater penetration is obtained. Conversely, on the decreasing-temperature slope portion of the transient curve the ξ thickness is much greater than the equivalent isothermally-produced layer would have been, and the rate is lower.

Estimation of Embrittlement Caused by Specific LOCA Transients

Examples are given in this section of calculated estimates of ξ penetration and embrittlement for several hypothetical transients and for one published curve derived from calculations by a reactor vendor.⁴ The purpose of these is to provide insight into the time-temperature effects of steam exposure on the microstructure (ξ values) and mechanical properties (ZDT values) of Zircaloy cladding. We discussed earlier the technique for calculating ξ penetrations for any transient curve and, by using the assumptions of two-surface exposure and full wall thickness, estimating the embrittlement that would occur. The values of ξ for some of the transients represent refinements in our calculational methods and differ from values published elsewhere for the same curves. In general, the values of ξ were found to increase for the new calculations. The reason was discussed in the previous section.

It should again be emphasized that the ZDT values are calculated assuming full-wall thickness tubing oxidized on both surfaces. The embrittlement results derived from two-surface calculations are conservative when applied to unruptured tubing (no inner surface oxidation) and nonconservative in the vicinity of the cladding rupture point. The area around the rupture in a real tube will be more brittle than our results indicate because of wall thinning.

Figure 13 shows a set of four hypothetical curves which are mathematically identical except each curve differs 100°F from its nearest neighbors in temperature over a peak temperature range from 2300 to 2600°F.

⁴Public Service Company of New Hampshire, The United Illuminating Company, Seabrook Nuclear Station - Unit No. 1, Preliminary Safety Analysis Report, Vol. III, Section 14, Fig. 14.3-14.4, Docket 50-340.

Fig. 13. Exposure Temperature Versus Time for Four Hypothetical LOCA Transient Curves. These are identical in shape and vary only in elevation on the ordinate.

These curves were integrated with the results shown in Fig. 14. Cumulative values of ξ are plotted against transient time above 1600°F. Curve A, corresponding to a 2300°F peak-temperature transient, yielded a ξ value of 0.00875 cm. This was doubled to represent two-surface exposure and used to calculate on F_W value — for a PWR wall thickness of 0.0686 cm — of 0.745. This is equivalent to a ZDT value of approximately 0°F. Similarly, values of ξ and ZDT are shown for curves B, C, and D. Because of the rapid rise of ξ with time for curve D, F_W and ZDT were calculated for the time the transient reached its 2600°F peak temperature. At this point, 71 sec after the transient passed 1600°F, the calculated ZDT was 2100°F.

Figure 15 illustrates four hypothetical transients with time scales more appropriate to BWR's. The total ξ values are shown, and the cumulative values are plotted versus time in Fig. 16. The effects of transient curve shape on the integrated time-temperature relationship are not readily apparent from Figs. 13 through 16. However, if one studies the instantaneous rate of ξ penetration with time at a given temperature these effects become quite apparent. Figure 17 is a plot of ξ versus time (above 1600°F) for curves C and D from Fig. 13 and curve 4 from Fig. 15. Curves C and 4 peaked at 2500°F and curve D peaked at 2600°F. The fourth curve (2400°F isothermal) is calculated from Fig. 5 for specimens isothermally exposed to steam for various times at 2400°F. The

Fig. 14. Cumulative Values of ξ as a Function of Time Above 1600°F for the Four Curves in Fig. 13.

Fig. 15. Exposure Temperature Versus Time for Four Hypothetical LOCA Transient Curves with Time Scales that Approximate Estimated BWR Transient Times.

Fig. 16. Cumulative Values of ξ as a Function of Time Above 1600°F for the Four Curves in Fig. 15.

Fig. 17. Penetration (ξ) Versus Time Above 1600°F for Selected Transient Curves from Figs. 13 and 15 and for a Parabolic Curve Calculated from Fig. 5.

slopes of the curves at points at which the transients passed through 2400 and 2500°F are shown as short lines tangent to the curves. Figure 18 is a plot of these slopes, $\frac{d\xi}{dt}$, versus the instantaneous values of ξ at the times the cumulative curves passed through 2400 or 2500°F. It is seen that the rate of ξ penetration initially decreases very rapidly with ξ and then becomes more nearly constant for larger values of ξ . The difference in the two 2400°F curves is attributed to the probability that ξ penetration due to isothermal exposure is not parabolic for short exposure times, as we have drawn it in Fig. 17. Instead, its initial slope should more closely approximate the other 2400°F curve, and therefore cause the $\frac{d\xi}{dt}$ versus ξ curves for 2400°F in Fig. 18 to coincide.

Fig. 18. Slopes, $\frac{d\xi}{dt}$, Taken from the Curves in Fig. 17 Versus the Instantaneous Values of ξ at the Times the Cumulative Curves Passed Through 2400 or 2500°F.

Finally, curves are shown in Fig. 19 for transients with time scales more appropriate to PWR's. All of the curves are hypothetical except the one indicated as coming from the Seabrook Nuclear Station.⁵ We translated the Seabrook curve shape upward in temperature to peak at 2500°F and also extended it to longer times. These five curves were integrated, and ξ as a function of time is given in Fig. 20. The same observations hold for these transients as for the previous ones.

It was mentioned earlier that there is nonconservatism inherent in the embrittlement data presented in this report, with respect to the cladding near the rupture. Not only will the tubes oxidize on both inner and outer surfaces after rupture, but that oxidation will be working on a thinned wall. Figure 21 contains experimental points that represent maximum and minimum wall thicknesses found in BWR-size tubes following fuel rod failure burst tests. The thicknesses are plotted against the circumferential expansions obtained. It was mentioned earlier that the transformed β -phase is the only ductile material in

⁵Public Service Company of New Hampshire, The United Illuminating Company, Seabrook Nuclear Station - Unit No. 1, Preliminary Safety Analysis Report, Vol. III, Section 14, Fig. 14.3-14.14, Docket 50-340.

Fig. 19. Exposure Temperature Versus Time for Five LOCA Transient Curves with Time Scales More Appropriate to PWR's. One of the transients is taken from the literature and the others are hypothetical.

Fig. 20. Cumulative Values of ξ as a Function of Time Above 1600°F for the Five Curves in Fig. 19.

Fig. 21. Maximum and Minimum Wall Thicknesses Found in BWR-Size Tubes Following Fuel Rod Failure Burst Tests. These values are plotted against the circumferential expansions obtained in the tests.

the tube wall. It is obvious that, for a given LOCA exposure, the value of ξ will generally be independent of wall thickness and, therefore, the ductile β -phase will be thinner by the amount the wall has thinned. We have seen this in a TREAT reactor LOCA test.⁶ Our analysis, based on full-wall-thickness tubing, predicted room-temperature ductility, yet the cladding was completely brittle around the thinned rupture area. Geometrically speaking, the maximum decrease the thickest remaining wall section of the tube (that expanded 125%, in Fig. 21) received was 40%. It is calculated that, if this expanded tube were exposed to steam at 2500°F for 2 min, for example, the maximum remaining β thickness would be approximately 0.014 cm instead of the approximately 0.049 cm that

⁶R. A. Lorenz and G. W. Parker, Final Report on the Second Fuel Rod Failure Transient Test of a Zircaloy-Clad Fuel Rod Cluster in TREAT, ORNL-4710 (December 1971).

would remain if the wall were unthinned. Experimental data to quantitatively define the amount of embrittlement one could expect from the above example do not presently exist, nor do data for various wall thicknesses or for unequal amounts of oxidation on the two tubing surfaces. We have shown how ξ varies with steam exposure conditions and have rigorously defined embrittlement as a function of two-surface exposure, but we can only remain qualitative on the important points mentioned in the preceding sentence.

It seems worthwhile to discuss some of the embrittlement consequences that may be related to unequal oxidation of the two tubing surfaces. The tubes used in this study were uniformly oxidized on both surfaces under non-steam-limited conditions. The experimentally determined values of ξ on the inner and outer surfaces of each tube were almost identical. Therefore, the rate of encroachment of the brittle phases or, to use a more useful comparison, the rate of decrease of the ductile β -phase was a maximum. If oxidation had occurred on only one surface, the rate of decrease of the β -phase would have been approximately halved. Obviously, any combination of times for start of oxidation on the outer and the inner surfaces will give a different rate of decrease of the β -phase; the decrease will be a maximum for equal oxidation of both surfaces and a minimum for oxidation on the outer surface only.

CONCLUSIONS AND RECOMMENDATIONS

We have shown the effects of steam exposure at elevated temperatures on the microstructure and mechanical properties of Zircaloy cladding. Our results are based upon data obtained from experimental work performed as described in this report.

Basically, exposure of Zircaloy cladding to steam at temperatures of approximately 2300°F for times of less than 1 min yields cladding with good ductility remaining at room temperature. This statement is based upon exposure of full-wall-thickness tubing to steam on both the inner and outer surfaces. Transient curves reported by utilities in preliminary and final safety analysis reports (none of which showed LOCA transient peaks greater than about 2250°F) and which were calculated from

assumptions of the design-basis accident and ECCS effectiveness were all found to yield ZDT values well below room temperature by our method.

Exposure temperatures above 2300°F were found to produce greatly increased amounts of embrittlement with increasing exposure temperature. For example, cladding exposed to steam, again on both surfaces, at 2400 and 2500°F for 1 min would show zero ductility below approximately 150 and 500°F, respectively. Exposure for 2 min at those temperatures would yield ZDT's of approximately 400 and 1000°F, respectively.

Three factors are important in determining the amount of embrittlement a tube will exhibit. Two of these, time and temperature of exposure, we have combined in our integration method. Of the two, temperature is by far the more effective in promoting embrittlement. Reference to Fig. 12 shows the rapidly increasing penetration rate of the brittle phases into the tube wall above 2300°F. The third factor is wall thinning. Depending upon the internal pressure present in the fuel rods, the cladding will rupture at temperatures ranging from approximately 1000 to 2500°F (pressures from approximately 2000 to 50 psig, respectively). Such a rupture results in considerable wall thinning in its vicinity and exposes the inner cladding surface to steam attack. Our analysis is, therefore, qualitative and nonconservative near the rupture area. That is, the volume of metal in the tube wall near the rupture is subjected to greatly increased embrittlement due to the wall thinning — which decreases the distance (ξ) necessary for the brittle phases to penetrate to effect embrittlement — and to oxidation from both surfaces. These consequences are ameliorated to some extent by the fact that inner surface oxidation will probably be confined to short distances on each side of the rupture point by the lack of an abundant steam supply inside the fuel tube.

The following recommendations are given to point out areas of study needed to more fully resolve the remaining questions about embrittlement.

1. Isothermal exposure of Zircaloy cladding to steam on the outer surface only. Data obtained here could be analogous to those obtained by two-surface oxidation in this present study. Information obtained would provide the minimum embrittlement rates to be expected for given

LOCA transients and would provide baseline data for the following recommendation.

2. Nonuniform oxidation of the two surfaces during an experimental transient. This represents perhaps the most meaningful controlled procedure for simulating the effects of a major LOCA on a fuel rod. Suggested implementation includes a series of internally heated tubes run at various transient conditions. Either rupture could be induced during the transient so that oxidation commences on the inner surface sometime after it starts on the outer surface, or else steam could be introduced to the inside of the tube at a predetermined time in the transient. The former procedure provides information on the effects of wall thinning as well as on nonuniform oxidation. The latter provides more direct experimental control of oxidation conditions and with constant wall thickness being maintained.

Both items 1 and 2 require mechanical testing of ring specimens as described in the body of this report. Completion of the studies as outlined above would allow very meaningful analyses of core embrittlement to be obtained. Given a distribution of transient rates and steam flows throughout the reactor core during a postulated LOCA, one could assess the embrittlement of that core with far fewer assumptions than are now needed.

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APPENDIX A

APPENDIX A

A. W. Lemmon, Jr., edited a report ⁷ in 1957 in which studies of diffusion of oxygen in α - and β -Zircaloy 2 and Zircaloy 3 were discussed. This work has been cited in more recent studies dealing with oxidation and embrittlement of reactor fuel cladding. It is relevant at this time to discuss the method used to determine the diffusion coefficient for oxygen in α -Zircaloy and to relate that to the real case for oxygen diffusion into Zircaloy fuel cladding. According to Battelle Memorial Institute, a moving-boundary technique described by Jost⁸ was used. The displacement, ξ , of the phase boundary produced by the diffusion of a single species is proportional to the square root of the time of diffusion, or

$$\xi = 2\gamma \sqrt{D_{\alpha} t} , \quad (A1)$$

where

ξ = displacement of the phase boundary, cm,

γ = a dimensionless parameter characteristic of the system,

D_{α} = diffusion coefficient of oxygen in α -Zircaloy, and

t = time, sec.

The parameter, γ , is defined by the report as

$$\frac{C_S - C_{II,I}}{C_{II,I} - C_O} = \sqrt{\pi} \gamma e^{\gamma^2} \operatorname{erf} \gamma , \quad (A2)$$

where C_S , $C_{II,I}$, and C_O are concentrations of the diffusing species at the various phase boundaries shown in Fig. A-1(a). There is an error in Eq. (A2). According to Jost,⁸ the $\sqrt{\pi} \gamma$ portion of the equation should be $\sqrt{\pi} \gamma$. It is not known whether this is a misprint in the BMI report⁷

⁷A. W. Lemmon, Jr., Studies Relating to the Reaction Between Zirconium and Water at High Temperatures, Battelle Memorial Institute USAEC Report BMI-1154, pp. 4-8 (Jan. 3, 1957).

⁸W. Jost, Diffusion, pp. 69-72, Academic Press, New York, 1952.

Fig. A-1. (a) Oxygen Concentration Profile Corresponding to the Diffusion of Oxygen into Zircaloy With an Already Oxygen-Saturated β -Phase. (b) Same as (a), but with the β -phase unsaturated. Two gradients are present and the rate of movement of the ξ boundary is slower than in the (a) example.

or whether the equation was actually used as shown in Eq. (A2). Figure A-1 (a) illustrates the oxygen concentration gradient in the α -phase, and the oxygen-saturated β -phase, described by the correct form of Eq. (A2). The real situation for diffusion of oxygen into Zircaloy cladding is shown in Fig. A-1(b) and involves two gradients instead of one. The diffusion equation for this situation is also given by Jost:⁸

$$c_{II,I} - c_{I,II} = \frac{(c_s - c_{II,I})e^{-\gamma^2}}{\sqrt{\pi} \gamma \operatorname{erf} \gamma} - \frac{(c_{I,II} - c_o)e^{-\gamma^2}}{\sqrt{\pi\phi} \gamma [1 - \operatorname{erf}(\gamma\sqrt{\phi})]}, \quad (A3)$$

where c_s , $c_{I,II}$, $c_{II,I}$, and c_o are as shown in Fig. A-1(b) and γ is as defined for Eq. (A2). The parameter ϕ is the ratio of D_α to D_β . If c_o is set equal to $c_{I,II}$, Eq. (A3) reduces to the correct form of Eq. (A2), that is, with $\sqrt{\pi} \gamma$ instead of $\sqrt{\pi\phi} \gamma$. The outer oxide layer can be

assumed to be a source of oxygen for the diffusion process at higher temperatures.

Although it is not strictly legitimate to vary the quantities in Eq. (A3) parametrically, since it is an integral equation representing a semi-infinite medium, it is instructive to see what effect changes in C_O and temperature have on the value of γ . Accordingly, values of C_S , $C_{I,II}$, and $C_{II,I}$ at each temperature were read off of the zirconium-oxygen equilibrium phase diagram and values of ϕ were calculated from diffusion coefficient equations given in the BMI report.⁷ Figure A-2 illustrates the variation of γ with C_O and temperature. The upper values of C_O are limited to the values of $C_{I,II}$ at any specific temperature. Therefore, the curve C-F represents the values of γ that should have been obtained through the use of Eq. (A2). We obtain a value of $\gamma = 0.81$ at 2200°F (1200°C) compared to BMI's $\gamma = 0.895$. This discrepancy can be explained several ways. We used atom fraction concentration values; BMI apparently used weight percent. The printing error in Eq. (A2) may be real. Nevertheless, it is seen that, at constant temperature, γ increases with C_O , reaching a maximum at $C_O = C_{I,II}$. Referring to Eq. (A1), it is seen that D_α is constant with temperature so that, at any given time, t , ξ varies directly with the value of γ . $C_O = C_{I,II}$, therefore, represents the condition for maximum displacement rate of the oxygen-stabilized α -phase into the cladding wall. This can be visualized by observing in Fig. A-1(a) that any oxygen that diffuses to the phase boundary is immediately used to transform β to α and thus increase ξ . In the case of two gradients, Fig. A-1(b), oxygen diffusing to the phase boundary may form new α or it may move through into the β -phase due to the second gradient.

The consequences of this discussion are important because of the implied increase in rate of boundary displacement and, therefore, rate of embrittlement that should occur as the oxygen concentration of the β -phase increases. Such increases can occur in specimens exposed to long times and high temperatures so that C_O increases or they can occur when the tube wall thins during rupture. The decreased diffusion path should increase the rate at which oxygen is supplied to increase C_O .

Fig. A-2. Three-Dimensional Graph Showing the Variation of γ , the Dimensionless Parameter Characteristic of the Diffusion System, as a Function of Temperature and C_0 , the Minimum Oxygen Concentration in the Tube Wall.

APPENDIX B

APPENDIX B

It is possible to calculate the approximate amounts of metal thickness converted to oxide (MTCTO) from measurements taken on our oxidized specimens. Details of these calculations are included here to show our approach, since there are a number of ways to perform this calculation.

The densities of zirconium and ZrO_2 used were 6.50 g/cm^3 and 6.10 g/cm^3 , respectively. The ZrO_2 density was multiplied by the mole fraction (0.74) of zirconium in ZrO_2 to get a value of 4.51 for the "effective" density of ZrO_2 . Measurements of total wall thickness and the oxide thickness were taken on the oxidized specimens.

The thickness of zirconium converted to ZrO_2 , per side, was calculated by:

$$\text{Thickness zirconium converted: } T_{\text{conv}} = T_{\text{oxide}} \left(\frac{4.51}{6.50} \right), \quad (\text{B1})$$

where T is thickness.

The amount of unconverted zirconium remaining, including both α - and β -phases, is given by:

$$\text{Thickness of unconverted zirconium: } T_{\alpha+\beta} = T_{\text{wall}} - 2(T_{\text{oxide}}). \quad (\text{B2})$$

The original wall thickness was obtained by:

$$T_{\text{orig}} = T_{\alpha+\beta} + 2(T_{\text{conv}}). \quad (\text{B3})$$

Finally, the percentage of original metal thickness converted to oxide is:

$$\% \text{ MTCTO} = \left(1 - \frac{T_{\alpha+\beta}}{T_{\text{orig}}} \right) 100. \quad (\text{B4})$$

No adjustment is made for thickness changes from lattice expansion due to oxygen dissolved in the α - and β -zirconium.

Our results are shown in Fig. B-1 in which the percent metal thickness converted to oxide is plotted as a function of fraction of wall

Fig. B-1. Percent Metal Thickness Converted to Oxide Plotted Against the Fraction of Wall Consisting of β -Phase (F_W).

consisting of β -phase (F_W). It is seen that conversion of 26% of the wall thickness to oxide (13% per side) would result in total elimination of the β -phase by the encroaching α -phases ($F_W = 0$). The linear equation describing these data is given approximately by:

$$\% \text{ MTCTO} = 26 - 26F_W . \quad (\text{B5})$$

The actual coefficients, obtained from least squares analysis, were 26.09 and 26.26 for Eq. (B5).

Of further interest are plots correlating various dimensional changes with amounts of oxidation. Figure B-2 is a plot of total wall thickness as a function of F_W showing the linear increase in wall thickness with increasing oxidation (decreasing F_W). Taken in conjunction with a back-calculated average value for original wall thickness of 0.0267 in. (standard deviation = 0.000279), it is seen that maximum wall thickness growth ($F_W = 0$) is 0.0028 in. The fractions of ξ consisting

Fig. B-2. Calculated Total Wall Thickness as a Function of Amount of Oxidation, Expressed in Terms of F_W .

of ZrO_2 and α are shown in Fig. B-3 as functions of ξ . The average measured fractions of ZrO_2 and α are 0.422 ($\sigma = 0.0410$) and 0.578 ($\sigma = 0.0408$), respectively. The plots in Fig. B-3 show that the linear least squares fits of the data (coefficient of correlation = -0.421 and $+0.401$, respectively; not good, but sufficient to show trend) indicate an increasing proportion of α -phase in ξ with increasing oxidation.

It was possible to check the internal consistency of the data by assuming that a specimen was oxidized until the midwall β -phase was eliminated ($F_W = 0$). Values of maximum wall thickness, fractions of ZrO_2 and α -phase thicknesses, and percent metal thickness converted to oxide were obtained by extrapolation, using Figs. B-2, B-3, and B-1, respectively. The calculated total thickness of the combined ZrO_2 and α -phases was found to be 0.0287 in., which compared well with the extrapolated value of 0.0295 in. total wall thickness.

The trend of an increasing proportion of α -phase in ξ discussed earlier in this appendix and the 0.0008-in. discrepancy of closure calculated above do not disagree with the increase in the rate of α -phase boundary movement implied in Appendix A. In a real, rather than extrapolated, situation, it would be expected that the 0.0008 in. of β -phase would be eliminated by the increased boundary movement.

Fig. B-3. Fraction of ξ Consisting of ZrO_2 or α -Phase as a Function of ξ .

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