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ZIRCALOY-2 PRESSURE TUBING

DECEMBER 1968

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DEVELOPMENT REPORT**

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ZIRCALOY-2 PRESSURE TUBING

Edited by

R. L. Knecht and P. J. Pankaskie

Metallurgy Research Department
Chemistry and Metallurgy Division

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ZIRCALOY-2 PRESSURE TUBING

PREFACE

The U.S. Heavy Water Reactor Program was reoriented in mid-1967 from a reactor development-construction project to a viable research and development program. The objective of the ongoing program was to establish and maintain a technology base that would allow the U.S., without committing to reactor construction, to retain the option to build an advanced converter based on water reactor technology. A program office (HWRPO) was established at Battelle-Northwest to assist the USAEC in defining, administering, and integrating this multitask base R&D program.

One of the first steps in establishing the base HWR program was to define the present status of technology in each major area of the various HWR systems. For the pressure tube type HWR systems, HWRPO requested the several R&D components of Pacific Northwest Laboratory (PNL) to collect and evaluate the adequacy of available technology on Zircaloy-2 pressure tubing. PNL was selected because of their extensive experience in this area including

- The design, fabrication, inspection of some 1200 pressure tubes for N-Reactor and the Plutonium Recycle Test Reactor,
- Continuing responsibilities in operational surveillance of the pressure tubes in those reactors,
- Continuing development work on advanced pressure tubing under the USAEC/AECL Cooperative Program.

A brief discussion of the direction and orientation of pressure tube work under the HWR program may help in understanding the constraints placed on PNL's review of pressure tube technology. The major objective of the pressure

tube program is to formulate a modest program lasting 2 to 4 years, the technical bases for the design and reliable operation of pressure tubes. While the tube thus developed is not expected to be the optimum in reliability or lifetime, its completion must be assured so that pressure tubes as a major HWR component will offer little deterrent to the commitment to build a HWR in the U.S. As this primary program moves toward its objective, secondary efforts will be underway on incentives for and development of advanced pressure tube designs and materials.

For the design of the primary pressure tube, no engineered alternative to a "hot-walled" tube is available. Various insulated and composite pressure tube concepts have been investigated in the laboratory, but few have progressed beyond a feasibility decision and none significantly into engineering development. The primary HWR pressure tube, therefore, will be a hot-walled type.

In the selection of zirconium alloy, a substantial technical appraisal and a decision are necessary. The Zircaloy(s) are undoubtedly the most extensively developed zirconium alloy(s). More is known about Zircaloy-2 as an engineering material of construction than any other zirconium alloy. This is particularly true of the breadth and depth of irradiation experience in terms of nuclear environments, physical sizes and shapes, number of specimens and neutron fluence. Tentative ASTM specifications for Zircaloy-2 are available. Zircaloy-2 products are in continuous commercial production.

In terms of component development, again, Zircaloy is by far the most widely used material. There are presently about 1600 Zircaloy-2 pressure tubes in operating HWR's and numbers of these tubes have accrued up to 6 years of service in prototypic HWR environments. Much is being learned from these

tubes and all evidence indicates they can be expected to operate satisfactorily for another 6 years. For the next most highly developed zirconium alloy, Zr-2.5% Nb, there are about 20 pressure tubes in operating reactors with several tubes at 1 to 2 year equivalent HWR exposures. Based on these early data, the Zr-Nb tubes are exhibiting even better behavior than the Zircaloy-2 tubes. Other zirconium alloys appear quite promising for pressure tube applications, but tests of these alloys have not progressed very far beyond the uniaxial coupon phase.

These fairly substantial differences in "engineered technology" between Zircaloy-2 and other zirconium alloys, notably Zr-Nb, could be offset by a significant probability that Zr-Nb would exhibit a longer reactor life. At this time, however, pressure tube life itself and the effects of various metallurgical and nuclear parameters have not been defined. For the probable limits to pressure tube life, flaw growth, hydrogen-affected fracture toughness, or creep, Zr-Nb appears to be equivalent to or better than Zircaloy-2. One of the uncertainties with Zr-Nb, however, is the metallurgical stability of its two-phase structure under long term neutron irradiation. Neutron-induced overaging or reversion are typically deleterious to stable creep rates and can lead to "premature" failure.

When the uncertainties of the other zirconium alloys were compared to the assurance qualification of the program objective, the decision was that HWRP continue to build on the available Zircaloy-2 base. Accordingly, PNL was requested to concentrate on Zircaloy-2 pressure tube technology in their status review. Data on other alloys and from Zircaloy-2 uniaxial specimens were to be included only to fill a gap in Zircaloy-2 pressure tube technology or, as a brief comparison,

to indicate the incentives possible in other alloys. In the interest of a timely completion, PNL's review was not to be exhaustive in the sense of collecting and reviewing all the data available in each subject area. Thus, the absence of any particular work from this status report is a result of selection and implies no decision as to technical significance or applicability. Finally, PNL's objective was to collect and review the available technology. In-depth analysis and interpretation would be the responsibility of future task groups.

ZIRCALOY-2 PRESSURE TUBING

ABSTRACT

A review of the present state of Zircaloy-2 heavy water reactor pressure tube technology with respect to design, specifications, production, use, operational standards and surveillance activity reveals an industry that has matured in a short span of years. There are still significant knowledge gaps particularly regarding the long term behavior of Zircaloy-2 and Zircaloy-2 pressure tubes under a variety of reactor service conditions.

ZIRCALOY-2 PRESSURE TUBING

Edited by

R. L. Knecht and P. J. Pankaskie

1.0 INTRODUCTION AND GENERAL COMMENTS

Within the United States and Canada, there are several water cooled thermal reactors that use pressure tubes as fuel channels in the core section of the primary coolant circuit. In the operating reactors almost all pressure tubes were made of Zircaloy-2, with perhaps a dozen or so experimental pressure tubes of Zr-2.5 wt% Nb alloy in several of these reactors. In existing reactors there are roughly 1600 pressure tubes ranging in size from about 2 1/2 in. ID to about 5 in. ID and wall thickness ranging from about 0.15 to about 0.27 in. Lengths range from about 10 to 50 ft. All tubes are seamless and were produced by hot extrusion and cold finishing by either tube reducing or drawing methods. Final material condition of the pressure tubes ranges from full annealed to about a 45% cold-worked temper.

In general, the design of the pressure tubes for all existing reactors was based on the intent of either Section III or VIII of the ASME code. Allowable working stresses were derived from material properties data obtained from uniaxial coupon type testing programs. For engineering application such data are best used for comparative purposes and subsequent experience in pressure tube evaluation programs has shown that such data have limited application to the behavior of thick-walled pressure tubing. This is especially so for material which exhibits a high degree of anisotropy in its crystallographic and mechanical properties, as does zirconium. Hence, the design of pressure tubes now in service appears to be somewhat conservative. However, considering that Zircaloy-2 had

not previously been used on any large scale, such conservatism was perhaps justified. For future reactors, efforts should be directed to maximum safe utilization of the properties of prototypic pressure tubing.

Of the 1600 or so pressure tubes now in reactor service, only a relatively few have been in service for as long as about 6 years. In general, the performance of the Zircaloy-2 pressure tubes has been good. In the PRTR, where maximum service has been experienced, there was some difficulty with fretting corrosion. Small changes in fuel element design and reactor operation have minimized this problem. These tubes have also been decontaminated twice without especial difficulty or ill effect. Neutron exposure ($E > 1 \text{ MeV}$) of the tubes with maximum service is now about 10^{21} nvt or slightly greater.

Substantial work has been done on mechanical and structural properties of pressure tubing. These efforts have been directed toward assessing the effects of temperature, cold-work, hydriding and neutron irradiation on burst strength, ductility, fatigue and fracture, and creep behavior. Data so far available suggest that the design basis used for the existing reactors may have been conservative. However, since service experience still is so short term, it would be difficult now to assure existing pressure tube designs for long term nuclear service, i.e., 30 years. Any irradiation effects data now in existence fall short of the neutron exposure anticipated over a 30 year period in a power reactor.

The purpose of this report has been to bring into sharper focus, the current state of Zircaloy-2 pressure tube technology.

2.0 SUMMARY AND CONCLUSIONS

The status of Zircaloy-2 pressure tube technology is presented. Tube design, specification, production, use and operational standards and surveillance activity are reviewed and discussed. Included are the results of supporting research and development work principally relating to strength and fracture properties as determined on tube and coupon type specimens in both the unirradiated and irradiated conditions. Information on other pressure tube materials, principally Zr-2.5 wt% Nb, is given.

Prior to the design and construction of the Plutonium Recycle Test Reactor (PRTR), there were few, if any, investigations of the properties and behaviors of zirconium alloy pressure tubes. Since we are primarily interested in the behavior of thick-walled tubing, little reference is made to material R&D efforts prior to about 1959-1960. Tables of data have not been included in this report but are referenced for the interested reader. In some cases, data have not been formally published and are available only from individual investigators. (These references are cited as Unpublished Data, personal communications.) The bulk of data regarding pressure tubes was derived from R&D efforts at Hanford in the U.S., from Chalk River National Laboratories in Canada, and from the Reactor Materials Laboratory in England. These laboratories currently have significant R&D efforts with zirconium alloy pressure tubing.

Based on this State-of-the-Art review, Zircaloy-2 is an excellent candidate material for pressure tubes in water-cooled thermal power reactors. Significant economic gains in the future may be possible through tube design using data being developed on strength properties of tubes under irradiation. Tube specifications must continually be reviewed and

updated to keep pace with technology and to thus maintain quality assurance. Where long term service is economically mandatory, some provisions for operation surveillance appear necessary at least until additional data and experience are acquired. While some reactor designs may require some highly specific data, the general area of R&D work should include 1) creep, flaw growth, and crack propagation behavior under prototypic biaxial stress ratios, and 2) corrosion and hydrogen uptake where behavior may be affected singly or in some combination by fabrication and texture, and by neutron flux and exposure. The magnitude of the irradiation problem yet to be evaluated can be appreciated from the fact that a 30 year exposure is about eight times that of the highest exposed pressure tube to date.

During that interim period when the above data are being acquired, the reactor operator will likely need to undertake some pressure tube surveillance. For this job, additional surveillance R&D appears to be warranted. Based on experience to date, a surveillance effort would include tube surface inspections, some check for dimensional changes and a nondestructive method for detecting size and location of subsurface flaws and extremely narrow surface cracks. Technology exists for doing these tasks; however, current instrumentation is too delicate and operation is rather complicated for occasional use by regular personnel in reactor operating organizations. Hence, some real beneficial R&D effort can be spent in appropriate instrument development.

In the design, construction and operation of nuclear power reactors, considerable emphasis is justly placed on safety and identification of the "maximum credible incident." However, there appears to be considerably less emphasis on slightly "off-standard" conditions that may occur at random intervals

during the lifetime of the reactor. Such conditions, of themselves, may not pose any safety hazard but may diminish the life of a pressure tube. Not only reactor operations, but also many experimental programs neglect reliability analysis. Application of reliability and sensitivity analysis to data from mechanical property R&D and in-reactor surveillance can and should provide valuable feedback into the direction of these efforts.

3.0 DESIGN BASIS

T. R. Ostrom and R. J. Evans

The zirconium alloy reactor pressure tubes in all Canadian Power reactors,⁽¹⁾ the Steam Generating Heavy Water Reactor (SGHWR) in the United Kingdom (U.K.),⁽²⁾ and the Carolinas-Virginia Tube Reactor (CVTR)⁽³⁾ in the U.S. were designed according to the intent of the ASME pressure vessel code. The applicable sections of the code were either Appendix Q of Section VIII⁽⁴⁾ for nonferrous metals or the relatively new Section III⁽⁵⁾ for nuclear applications. This code sets the maximum allowable stress at the lowest of the following criteria at design operating temperature:

- One-fourth of the ultimate strength (1/3 in Section III).
- Two-thirds of the yield strength.
- The stress to produce a secondary creep rate of 0.1% in 10,000 hr.
- The 100,000 hr rupture stress.

At an operating temperature of approximately 300 °C, the limiting criterion is the ultimate tensile strength. The Atomic Energy Control Board (Canada) has allowed the loop tubes at Chalk River to be designed to 1/2.3 the ultimate tensile strength rather than the 1/3 given in Section III or the 1/4 given in Section VIII. This allowance is due primarily to the experimental nature of the loop tubes and to the high frequency of tube inspection.⁽¹⁾ The other design criteria of Sections III and VIII were applied.

Since zirconium alloys are not coded materials, the pressure tubes were designed according to the intent of the code. By precedent, uniaxial, unirradiated material strength was used in applying the limiting code criteria with no limitations or allowances provided for the effects of neutron irradiation.

The limiting design criteria used for the PRTR and N-Reactor pressure tubes were based on the stress that would produce no more than 1% creep during the life of the tubes (PRTR tube life: 10 years; N-Reactor tube life: 20 years).⁽⁶⁻¹⁰⁾ At the time of these designs, it was not known whether Zircaloy-2 strengthening imparted by cold-work would be lost by annealing during long time service; thus, the strength properties determined on annealed uniaxial specimens were used. Conventional use of these data with the code criteria for creep and stress rupture was difficult. The creep rate was fairly insensitive to stress to near ultimate strength values. For the operating temperatures of interest, the hardening effect from strain aging made extrapolation of stress rupture data to a 100,000 hr rupture stress difficult. All long term uniaxial creep data for annealed Zr-2 showed that the strain at or before stress rupture exceeded 3%.⁽⁹⁾ After consideration of allowances for long term service and decreases in ductility from irradiation damage, a total allowable strain of 1% was selected as a conservative design criterion. Based on this criterion, the allowable design stress was determined and appropriate safety factors were applied in each case.^(8,10)

Increases in the strength properties of Zircaloy-2 from biaxial loading, from anisotropic strengthening, and from radiation effects are very significant. Counteracting these effects is accelerated strain of tubes under irradiation from an as yet not well understood phenomenon thought to be accelerated creep, radiation induced growth, or a manifestation of superplasticity. If appropriate design allowances can be made for increased strength properties, it would represent a considerable savings in tube costs and in reactor space and neutron economics. Experience and data on the biaxial and anisotropic strengthening of tubing are fairly well established

(see Section 5) and this information indicates that biaxial tube strengths 40% greater than uniaxial strengths may be obtained. For example, Aungst⁽¹¹⁾ has done biaxial stress rupture testing of N-Reactor tube specimens and concludes: "The design for Zircaloy-2 pressure tubing will be very conservative if it is based on creep data derived from uniaxial tests performed on strip material. A creep rate compatible with 1% strain in 20 years is achieved in a biaxial test of tube specimens at a stress 60% higher than in uniaxial tests of strip."

The irradiation strengthening of Zircaloy-2 is not well developed and that information available has to be qualified from a design viewpoint (Section 6 and Section 7). For example, information is available on uniaxial tests but the data cannot be correlated to tube experience. Also, much of the tube data have been gathered on irradiated tube sections out-of-reactor and, thus, out-of-flux. However, current programs at BNW and Chalk River are establishing creep rates of tubes in situ in PRTR, NRU and NRX reactors.

This specification does not stipulate the percent cold-work in the finished tube, the methods by which dimensions will be checked, the surface roughness, or detailed welding and welding inspection procedures.

The development of purchase specifications for the N-Reactor and PRTR tubes preceded and actually helped provide a basis for the ASTM specifications. They included all items covered by ASTM B-353 plus the items listed above which are not included in the ASTM specifications.^(2,3) In general, the Canadian tubes were ordered at later dates and the ASTM B-350 specification for ingots and parts of the ASTM B-353 specification for tubes are used.

Thus, although the specifications of Zircaloy-2 pressure tubes for the various reactors have extensive similarities, a common basic tube specification is not in use. Also, an important element that has not been specified, but which can be specified in future tube orders, is a specific minimum tube strength increase from crystallographic orientation, grain size and other effects.

4.2 FABRICATION

Zircaloy-2, Zircaloy-4, Zr-2.5 wt% Nb and Ohzennite alloy pressure tubes have been successfully fabricated. The procedures and industrial capabilities for producing zirconium base alloy pressure tubes were largely developed by Hanford personnel working with the tubing fabricators during the procurement of the PRTR⁽⁴⁾ and N-Reactor pressure tubes.⁽³⁾ Initial tubes were of poor structural, surface and dimensional quality and they reflected the underdeveloped state of fabrication technology for this hexagonal close packed metal. Development efforts were required in the following areas:

- Metal forming and annealing
- Extrusion temperatures, ratios, lubricants and tools

- Drawing and tube reducing methods, lubricants and tools
- Metal conditioning and
- Quality control.

The PRTR tapered tube⁽⁴⁾ which illustrates a wide cross section of fabrication (Figure 4.1) was fabricated from double vacuum arc melted material by the following major steps:

- Ingot forging and subsequent boring or piercing to obtain the extrusion billets.
- Hot extrusion of copper clad and steel interlined billets.
- Tube reducing in two steps to produce a tube with a constant outside diameter but with a thick wall length, and a thin wall length.
- Vacuum anneal of all the thick wall length (for tapering) and part of the adjoining thin wall length.
- Tube reducing the thick wall length of the tube to provide a taper and small diameter section.
- Flange attachment by a thread connection and seal welding.

The processing resulted in a tube with a variety of metallurgical conditions: the flange is seal welded; the large diameter section has a 45% cold-work structure which transitions to a vacuum annealed structure about 4 ft above the taper; the taper section increases from 0% cold-work at the top to 55% at the bottom; and the small diameter has a 55% cold-worked structure.

The length of the N-Reactor tubes, 50 ft, the diameter of the SGHWR tubes (5 1/8 in. OD), and the combination of straight and tapered portions of the PRTR tube are all good evidence of the wide range of industrial capabilities in the U.S. for producing pressure tubes. Also, additional industry capabilities have been demonstrated by contracts for various shapes of zirconium alloy products; hexagonal and square tubes have been

FIGURE 4.1. PRTR Zircaloy-2 Pressure Tube

extruded from round billets and coextrusions of Zr-2.5 wt% Nb with Zircaloy-2 clad to the inside and outside surfaces have been made.⁽⁵⁾

Thus, good quality tubes are now being produced but it is not a routine mass production process. Each tube is still produced as an individual item primarily due to the exacting dimensions and quality requirements demanded by reactor designers. Although several companies are capable of producing zirconium tubes, only Chase Brass and Copper and Harvey Aluminum are now actively engaged in zirconium pressure tube production in the U.S.

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5.0 STRESS RELATED PROPERTIES OF PRESSURE TUBING

T. R. Ostrom and R. J. Evans

5.1 CREEP AND STRESS RUPTURE

The only reports found on stress rupture testing of Zircaloy-2 in tubular geometries were those from Hanford. Data have been obtained from annealed PRTR tubing,^(1,2) drawn N-Reactor tubing of 18 and 30% cold-work, drawn KER loop tubing of 30% cold-work, and tube reduced N-Reactor tubing of 30 to 35% cold-work.^(3,4) These data show that 30% cold-worked N-Reactor and KER tubing behaved similarly and had better creep resistance than the 18% cold-worked material. The 35% cold-worked tube reduced material performed better than the 30% cold-worked drawn material. These differences were greater than could be explained by the 5% increase in cold-work and are now attributed to fabrication texture effects. Creep strength of annealed PRTR tube sections is less than the cold-worked material, but when creep strength is plotted as percent of burst strength versus time to rupture at 550 °F, there is good agreement with cold-worked data.

There were a few tests by Aungst and Evans^(3,4) which investigated the effect of autoclave treatment on the creep properties of tubing. Autoclaving, or the equivalent heat treatment, produced a decrease in primary creep strains, decreased the secondary creep rate, and increased the time to rupture. The same general effect was found by Knecht and Pankaskie⁽²⁾ in their comparisons of annealed tensile specimens from autoclaved PRTR tubing and unautoclaved Zircaloy-2 strip. Their work was done at 550 °F and at stresses from 12,500 to 20,000 psi. This work indicated that all the primary creep had occurred within about 50 hr and that for strip, autoclaving decreased primary creep by about 2 1/2

times for a stress of 12,000 psi and by about 4 1/2 times for a stress of 20,000 psi. The secondary creep rate, within measurement accuracy, appeared to be about the same.

Stress rupture testing on tubular specimens at Hanford has shown that a stress of approximately 50,000 psi at 300 °C will result in a total strain of 1% in 20 years, whereas for strip material a stress of 30,000 psi at 300 °C would result in the same 1% strain in 20 years.⁽³⁾

5.2 BURST STRENGTH AND DUCTILITY

Information is available on burst strength and ductility versus temperature for annealed Zircaloy-2 and Zircaloy-2 with 18, 30, 35, and 45% cold-work in the temperature range from room temperature to 300 °C.^(3,5,6) There are also data published by Watkins and Cockaday⁽⁷⁾ for 300 °C burst tests on drawn tubing of 20 and 30% cold work. These data are all presented in Figure 5.1; they indicate that for all test temperatures, the strength of 45% cold-worked tubing is greater by roughly 15,000 psi than that of fully annealed pressure tubing. The percent circumferential elongation is fairly high on all specimens but there does not seem to be any obvious dependence on the percent of cold-work.

Comparisons with data obtained from tensile coupons (see Section 7.0) will show that the strength of Zircaloy-2 in tubular geometries is significantly greater than predicted by the Von Mises-Hencky theory.

5.3 FRACTURE AND FATIGUE PROPERTIES

Crack propagation information is available on tubing from the PRTR,^(8,9) KER loop tubes, N-Reactor,⁽¹⁰⁾ and SGHWR.⁽¹¹⁾

FIGURE 5.1. Burst Strength and Circumferential Elongation of Zircaloy-2 Pressure Tubing and Its Variation with Temperature

This includes failure stress versus slot length* at room temperature and some information on elevated temperature slot tests on all but the KER loop tubing. The room temperature data (Figure 5.2) in the failure stress area below 25,000 psi hoop stress show fair correlation for failure stress versus slot length for the PRTR, KER loop and SGHWR tubes. The N-Reactor stress values were about 50% higher than the other three kinds of tubing. No conclusions are drawn on the effect of cold-work on the room temperature crack propagation tests because of the dissimilarity of the data received from the KER loop tubes, SGHWR tubes, and the N-Reactor tubes that all had 30% cold-work. Also, the tests on the PRTR tubing showed no difference in the crack propagation tests between the annealed and 45% cold-worked sections.

Data on elevated temperature crack propagation tests (Figure 5.3) show a decrease in hoop stress at failure with increasing temperature. The change in strength becomes less as the slot length increases. However, for the longer crack lengths, the strength is nearly constant with increasing temperature. This means that room temperature tests of hoop stress at failure versus slot length give a fair indication of the critical crack length at operating temperature as well as at room temperature.

The crack arrest temperature for both the NPR and PRTR pressure tubing were found to be between 50 and 150 °C.^(9,12) On the specimens that were tested below the crack arrest temperature, the tube failed full length when the 20% wall

* The slot test specimens reported in the Hanford data were made by milling a slot 80% through the tube wall with a 1/16 in. end mill. The slot test specimens run on SGHWR tubing were made by milling a slot through the full wall thickness.

FIGURE 5.2. Failure Stress Versus Slot Length at Room Temperature for PRTR, KER Loop, N-Reactor, and SGHWR Pressure Tubes

FIGURE 5.3. Hoop Stress at Failure Versus Test Temperature for PRTR, SGHWR, and N-Reactor Tubes with Different Slot Lengths

thickness web failed. On tests above the crack arrest temperature, crack propagation either does not occur or occurs only by ductile tearing.

High cycle fatigue test data is available from two N-Reactor samples of Zircaloy-2 pressure tubing,⁽¹³⁾ both of which indicated that the stress at failure was reduced from the value that was predicted from the slot length versus failure stress curve for N-Reactor tubing with milled slots. This decrease in strength was probably due to a combination of fatigue, work hardening of the material and increase in the sharpness of the crack. More work is needed to evaluate the effects of failure by cracks produced by fatigue and the interaction of multiple defects in tube failure.

Cracks electrojetted into the outside surface of the tube to depths equal to 40 and 75% of the wall thickness took 80,000 and 12,000 pressurization cycles, respectively, at 20,000 psi to penetrate the wall. The crack length on the outside surface when the cracks penetrated the wall was 0.6 in. and both tests took about an additional 25,000 cycles to grow to a length of 2.25 in. when they both propagated full length. Leak tests performed in both tests showed that at an outside surface crack length of about 1.2 in., the leak rate at about 500 psi internal pressure (2000 psi hoop stress) was 2 gal/hr, which was equal to the capacity of the pump. This test indicated that a crack would leak before failure.

More work is needed on hydrided and unhydrided tubular specimens at room and elevated temperatures to determine the differences between tube crack propagation characteristics when milled slots are used and when the "worst case" fatigue generated cracks are used.

5.4 EFFECTS OF FABRICATION

The final forming step of recently produced pressure tubing has been drawing and although no Zircaloy-2 reactor pressure tubes are presently being produced by the tube-reducing method used for the PRTR and some of the N-Reactor tubes, the differences in mechanical properties and subsequent service performance between tube-reduced and drawn tubing can provide a valuable insight on the effects of fabrication to future reactor designers.

Aungst has worked with the three types of N-Reactor tubes (35% tube-reduced, 18% cold drawn, and 30% cold drawn). His findings indicate that tube-reduced tubing has an ultimate burst strength slightly superior to drawn tubing and has significantly superior 300 °C stress rupture properties up to 10,000 hr.⁽³⁾ Accelerated stress rupture tests using higher temperatures and extrapolating 300 °C curves indicate that 20 year stress rupture strengths would not be significantly different between tube-reduced and drawn tubing.

Aungst's crack propagation studies⁽¹⁰⁾ using slotted tubes has indicated that the 30% drawn tubing is stronger than tube-reduced tubing. When hydrided tubing with short slots was tested at room temperature,⁽¹⁴⁾ the 30% drawn tubing was nearly twice as strong as the tube-reduced, but with 3 in. slots the tube-reduced material was significantly stronger than the drawn tubing. Work by Ostrom⁽⁹⁾ with PRTR annealed tubing sections has shown no lowering of the failure stress in slot tests with additions of hydrogen.

Discussions with Cheadle⁽¹⁵⁾ indicate that the different production methods cause a wide variation in the texture of the material and that the variation in mechanical properties can be related to this difference in texture. His findings

conclude that based on ultimate tensile strength, drawn tubing has a texture superior to tube-reduced tubing for use as reactor pressure tubes.

Apparently, there is a need to apply a systematic approach to existing data to understand the effect of methods of fabrication on the mechanical properties of zirconium-alloy pressure tubes. Since different test programs have used different sized tubing, an area of interest which might explain some of the apparent contradictions in findings would be the influence of geometry on the results of mechanical properties tests. For example, the PRTR tubes have a radius (r) to wall thickness (t) ratio of 10 to 1, while the N-Reactor r to t ratio is only 5 to 1 and typical sheath tubing may have a 15 to 1 or greater ratio.

Marshall's^(16,17) work with sheath tubing on orientation of hydrides indicates still another area of the effects of fabrication on tubing properties. His findings indicate preferred orientation of hydride platelets is a result of the interaction of two orienting forces: 1) a "natural" orientation habit originating during fabrication, and 2) a stress system acting on the zirconium matrix during platelet formation (stress orientation). He found that the susceptibility to stress orientation was very high for hot-reduced tubes with essentially 100% of hydrides reoriented at moderate stress levels; that for all cold-reduced tubing the stress orientation susceptibility was lower and very similar; and that cold-expanded tubing showed no stress orientation susceptibility at all. Cold reduction oriented the hydrides in a circumferential direction while free sinking operations increased the radial hydrides significantly.

Metallographic examinations of PRTR and N-Reactor tubing coupled with Marshall's work would indicate that the present

methods of pressure tube production (extrude and cold reduce) produce a texture for acceptable hydride orientations and that hot working and free sinking operations could produce undesirable hydride orientations in zirconium alloy pressure tubes.

Prior to installation in the PRTR, NPD (Canada) and N-Reactor, tubes were given an autoclave treatment. The autoclaving process, the final corrosion proof test, involves exposure to steam at 400 °C (425 °C for N-Reactor tubes) for several days. In addition to providing a protective oxide, it acts as a thermal stress-relieving treatment and provides long-term dimensional and mechanical stability for 300 °C operating conditions. Burst and tensile strengths are reduced from the as-drawn material, but primary and secondary creep strains are reduced 4 to 5 times by this treatment.⁽³⁾

5.5 HYDRIDE EFFECTS ON MECHANICAL PROPERTIES OF TUBES

The most significant effect of hydrogen on the mechanical properties of Zircaloy-2 is the embrittlement resulting from precipitation of zirconium hydride. Room temperature tests⁽¹⁴⁾ with hydrided Zircaloy-2 have indicated such a dramatic loss in ductility that considerable efforts by the U.S., Canada, and the U.K. have been expended to determine quantitatively the effects of hydrogen on the mechanical properties of Zircaloy-2 tube material.

With the exception of data based on corrosion coupons in the ETR, indicating the base metal of a Zircaloy-2 pressure tube has reached 100 ppm hydrogen,⁽¹⁸⁾ no cases of severe generalized hydriding have been found in zirconium alloy reactor pressure tubes. Localized^(8,19-21) hydriding has been found, but in no case has a pressure tube failed to operate satisfactorily due to any hydrogen buildup.

Sufficient work has been done to show that hydrogen which precipitates as zirconium hydride, and not hydrogen in solution, is the cause of embrittlement. Although the hydride precipitate may actually be in the form of a deuteride (i.e., in heavy water reactors), reported results generally assume no difference in mechanical properties between hydrided and deuterided material. Sawatzky⁽²²⁾ did show that equal atomic fractions of deuterium and hydrogen have equal effects on the impact strength of slowly cooled Zircaloy-2.

Specimens for testing are usually charged with hydrogen by a gas diffusion process at elevated temperature; however, Sexton⁽²³⁾ has reported a technique for low temperature hydriding.

Burst testing⁽¹⁰⁾ cold drawn N-Reactor tube specimens with increasing hydrogen levels to 275 ppm indicated no reduction in burst strength at either room temperature or 300 °C. The 275 ppm tests indicated zero ductility at room temperature and a slight loss in ductility at 300 °C.⁽³⁾ During burst tests of irradiated PRTR tubing, Fraser⁽⁸⁾ found increased hydrogen content in the region of the burst. He concluded that a combination of fret marks and hydrogen determined the burst origins but the reduction in strength due to fret marks and hydrogen was negligible. In addition, Sawatzky⁽²²⁾ and Irvin⁽²⁴⁾ found that tensile strength of Zircaloy-2 is unaffected by hydrogen up to 250 ppm.

Short term stress rupture tests at 300 °C with tubing hydrided to 275 ppm H₂ have indicated only a 5% loss in strength compared with unhydrided tubes.⁽²⁵⁾

Emphasis has been placed on determining the effects of hydrogen on the crack propagation of flawed tubing. Data is available on the combined effect of increasing amounts of

hydrogen to 400 ppm, flaw lengths to several inches and temperatures to 400 °C on the failure stress of Zircaloy-2 pressure tubing.^(7,26-30) Data is now being obtained on the combined effects of irradiation and hydrogen on flawed tubes.

There have been instances of hydrided burst specimens failing at very low stress levels. Examinations of the material usually revealed surface cracks and a concentrated hydride layer with evidence to indicate the cracks had occurred prior to the burst test. Investigations into the combined effects of multiple long shallow cracks in a brittle hydride layer on the strength of a pressure tube are just beginning at BNW and at Chalk River.

Zirconium hydrides which are oriented in a radial rather than circumferential or random orientation have a greater effect on reducing the ductility of tubes. The final cold working operation in current practices of tube production is apparently conducive to the preferred random or circumferential orientation. Marshall's^(16,17) work indicated that even circumferential hydrides can reorient themselves to a radial direction under service conditions if the material is above a threshold stress of 24,000 psi. Aungst,⁽¹⁴⁾ however, was not able to effect significant reorientation in the fine grained cold-worked pressure tubes at stresses up to 35,000 psi.

Data from the U.K.,^(7,26,28) Canada⁽²⁷⁾ and on the N-Reactor⁽¹⁴⁾ tubes have indicated significant losses of strength due to hydrides in burst tests of flawed tubing at room temperature. Similar tests of annealed PRTR tubing⁽⁹⁾ indicated no effect of hydrides on the flawed burst strength. At 300 °C, burst tests of tubing with hydrogen concentrations to 400 ppm indicate no strength loss due to hydrogen.

There are some differences in the results obtained from the U.K., Canada, and between the PRTR and N-Reactor tubes.

These differences may be due in part to experimental techniques (the U.K. and Canada use a gas pressurization system and PNL uses a liquid hydraulic system), and in part to geometry effects. All laboratories use tubing of different dimensions.

All of the test results indicate the pressure tubes in currently operating reactors could withstand a through-the-wall flaw several inches long and hydrogen concentrations of 250 to 400 ppm without failing.

Work with unhydrided Zircaloy-2 tubing⁽²⁵⁾ and Zr-2.5 wt% Nb tubing⁽²⁹⁾ to understand the mechanisms of fatigue crack growth is underway by fatiguing a flaw with cyclic internal pressures. This work also provides data as to the crack length at which it will leak and the critical length at failure. Preliminary results indicate that a crack will leak well before catastrophic failure. Future efforts will include the effects of hydriding.

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6.0 RADIATION EFFECTS ON PRESSURE TUBE PROPERTIES

T. R. Ostrom

6.1 CREEP AND STRESS RUPTURE

Creep of irradiated Zircaloy-2 pressure tubes is accelerated by the presence of a fast neutron flux above a threshold value and it is decreased when out of flux by the presence of irradiation damage sites that act as pinning sites for dislocations. Preliminary data show that in-reactor creep maintains a constant (flux dependent) secondary rate, while out of reactor tests show a continuously decreasing secondary rate. Postirradiation stress rupture tests show little change in primary creep and a faster decay of secondary creep rates when compared to tests run on as-received material.

For pressure tubing under neutron irradiation, relatively little creep and stress rupture data exist. Much of the in-reactor creep investigations of tubing is being done by AECL at the Chalk River National Laboratory. At CRNL, in-reactor creep data are being acquired from periodic diameter measurement of full-sized pressure tubes in situ and from special experiments with small diameter tubular test specimens. Additional creep work is being done at the PRTR under a USAEC/AECL cooperative program and under other AEC sponsorship.

Nine tubes in the NRU, NPD and PRTR are being measured to obtain in-reactor creep data. This program has been underway for several years. Data so far obtained from tests run in NRU and NPD will soon be published in the Journal of Nuclear Materials in a report, "The In-Reactor Creep of Cold-Worked Zircaloy-2 and Zirconium, 2-1/2 wt% Nb Pressure Tubes", authored by P. A. Ross-Ross and C. E. L. Hunt.

Three tubes in the NRU are stressed from 11,500 to 17,300 psi hoop at operating temperatures of 250 to 290 °C. Four tubes in NPD are stressed at 10,500 psi hoop at operating temperatures of 252 to 273 °C. Two tubes in the PRTR at Hanford are stressed at about 12,500 psi hoop at operating temperatures of 250 to 280 °C. Reactor exposure on these tubes ranges up to about 29,000 hr and approximately 10^{21} nvt ($E > 1$ MeV).⁽¹⁾

Special experiments with small diameter tubing are being done by E. F. Ibrahim.⁽²⁾ Tubular specimens 0.9 in. ID have been machined to provide six different hoop stresses, which range from 15,500 to 39,000 psi. The test assembly is constructed to expose six test specimens to a fast flux ($E > 1$ MeV), six specimens in a thermal neutron flux, and six specimens to little or no flux in the upper shielding region. Preliminary results on the secondary creep rate in flux agree with the finding of Ross-Ross⁽¹⁾ on full sized pressure tubes. No measurable difference in the thermal flux and out-of-flux specimens has been detected.⁽²⁾

At Hanford, there is a very modest effort in measuring pressure tubes. At the PRTR, pressure tubes are Pi taped (Pi taping consists of measuring the circumference of the tube to determine an average equivalent tube diameter) whenever a tube is temporarily or permanently removed from the reactor. So far, seven tubes have been Pi taped.⁽³⁾ While there is appreciable scatter in the data, the results are in general agreement with the finding of Ross-Ross. At the N-Reactor, an ID gage is available for tube diameter measurements but to date no measurements have been obtained.

In one of the Plutonium Production reactors, five thin wall (0.040 in. nominal) Zircaloy-2 coolant tubes have been measured after about 7 to 8 years of service. The hoop stress

in these tubes due to internal pressure was about 3000 psi and the coolant temperature is less than 100 °C. The diameter change of these tubes as determined from measurements at in-flux and out-of-flux positions was approximately 1/2%. The diameter profile curve had the same shape as the flux profile curve along the length of the tube and appears to be independent of the temperature along the length of the tube.⁽⁴⁾

From periodic measurements on pressure tubes in situ⁽¹⁾ and from Pi tape measurements at the PRTR⁽³⁾ and other measurements,⁽⁵⁾ it appears that creep behavior at reactor operating temperatures and relatively low hoop stress is proportional to fast flux intensity. For example, a graph of tube diameter versus tube length exhibits a cosine shape very similar to the flux intensity curve through the reactor core in a direction parallel to the axis of the pressure tubes. At least at the lower stresses (10,500 to ~17,000 psi hoop stress) measurable creep appears to occur only in the presence of a fast neutron flux ($E > 1$ MeV). Based on observations of Ross-Ross and Ibrahim,^(1,2) the following is tentatively concluded on the creep of Zircaloy-2 pressure tubes under irradiation:

- 1) creep is accelerated in a fast neutron environment;
- 2) secondary creep rate in-reactor tends to be constant, whereas, for out-of-reactor tests, the secondary creep rate would be continuously decreasing; and 3) secondary creep rate in-reactor appears to depend on the intensity of the neutron flux.

The experience with the thin-walled (0.040 in.) pressure tubes at low hoop stresses (~3000 psi) and low temperatures (less than 100 °C) confuses the in-reactor creep picture somewhat. In the absence of neutrons, creep at these hoop stresses and temperatures would not be expected. Hence, one must conclude that the effect of neutron radiation is indeed

large. Alternatively, one might speculate that at these low temperatures and stresses, the deformation observed may not be creep in the normal sense but a manifestation of radiation induced growth or superplasticity. Certainly more R&D effort is yet needed to fully assess the effects of neutron radiation on creep and stress rupture behavior. The threshold n_v for creep acceleration, the relationship between creep rates and n_v , the total attainable deformation as influenced by n_v and total fluence and the added⁽³⁾ effects of stress and temperature are of special importance in the design of efficient pressure tube for high temperature reactor service.

Some postirradiation stress rupture testing has been done at Hanford. Tests on KER loop tubes and N-Reactor pressure tubes show that prior irradiation damage reduces the secondary creep rates. Tests by Aungst⁽⁵⁾ have shown an increase in time to rupture for high stress, short term tests. A test by Aungst and Evans⁽⁶⁾ on KER loop tubing at 57,000 psi hoop at 300 °C continued under stress for 17,300 hr without failure. Total strain at 17,300 hr was 1.3% of which only 0.2% occurred in the last 10,000 hr. For this test, the average secondary creep rate was 2×10^{-6} %/hr. From a test of similar unirradiated pressure tubing, rupture occurred in 3024 hr at a hoop stress of 56,600 psi at 300 °C. Average secondary creep rate was 6.8×10^{-4} %/hr.

Based on the above data,^(5,6) a decrease in secondary creep rate is observed from postirradiation tests, whereas data from in-reactor tests^(1,2,3) show that creep rates are increased in a neutron irradiation environment. What may happen to stress rupture life is as yet unknown. Clearly, further in-reactor creep and stress rupture data are needed.

6.2 BURST STRENGTH AND DUCTILITY

The only reported data on burst strength, ductility, and crack propagation of irradiated tubular specimens of Zircaloy-2 are from Hanford. The AECL work in these fields has not yet been released for publication. The work at Hanford covers exposures up to about 3×10^{21} nvt ($E > 1$ MeV) and shows the effect of varying amounts of cold-work. Areas yet requiring investigation are the combined effect of hydrogen and irradiation and the effect of increasing nvt ($E > 1$ MeV) to about 10^{23} on the material properties. All post-service data need to be related to in-flux material properties.

The most reliable information on the effect of nvt on room temperature burst strength is available from PRTR process tubes.⁽⁷⁻¹¹⁾ There is also information on KER loop tubing of unknown exposure that shows an increase in burst strength from 5 to 20% for exposures up to 28 months.⁽¹²⁻¹⁵⁾ There is also limited information available on N-Reactor tubing in a confidential report, BNWL-CC-1092. The data from PRTR (Figure 6.1) show that the strength of an annealed tube is unaffected by exposures up to 5×10^{18} nvt ($E > 1$ MeV) and increases approximately linearly with the log of the exposure up to exposures of about 3×10^{21} nvt ($E > 1$ MeV). At exposures of 5×10^{20} nvt ($E > 1$ MeV), it is no longer possible to distinguish between 45% cold-worked material and annealed material, and their strength is still increasing linearly with log exposure to about 147,500 psi at 3×10^{21} nvt ($E > 1$ MeV).

The only available information on elevated temperature burst testing of pressure tubes of known exposure is on tubes removed from the PRTR reactor.⁽⁷⁻¹¹⁾ Some information exists on tubes removed from the KER loops with unknown exposures that show an increase in burst strength of about 20% above that of as-received tubing.⁽¹⁵⁾ The data from PRTR (Figure 6.1)

FIGURE 6.1. Hoop Strength at Failure Versus Fast Neutron Exposure of PRTR Zircaloy-2 Pressure Tubes

show results for room temperature tests similar to those at 300 °C. The burst stress has increased to 81,000 psi at 2.5×10^{21} nvt ($E > 1$ MeV) and appears to be still increasing with increased exposure. Existing information on the influence of test temperature on burst strength at constant or nearly constant exposure shows that the slope of the graph of hoop stress versus test temperature is constant.⁽⁷⁾ This is in fair agreement with the behavior of unirradiated material reported in Figure 6.1.

It has been found by Fraser⁽⁸⁾ and Ostrom⁽¹⁰⁾ that the ductility of irradiated material in both the annealed and cold-worked condition has decreased to a few percent at exposures ranging up to 3×10^{21} ($E > 1$ MeV). Aungst⁽¹⁴⁾ found a loss of ductility in KER loop material from the shield area of the K-Reactor that experienced almost no fast flux, which may indicate that part of this loss in ductility may be due to the time at temperature. Similar results were found by Aungst and Evans on the examination of an N-Reactor tube that is reported in a confidential document, BNWL-CC-1092. More work should be done to determine the effect of time at temperature on the loss of ductility in reactor tubes.

6.3 FRACTURE PROPERTIES

Information gathered from both KER loop tubes⁽¹⁵⁾ and PRTR process tubes⁽⁸⁾ indicates that there is little effect of irradiation on the crack propagation characteristics of Zircaloy-2 tubing at room temperature. A crack propagation test on a PRTR pressure tube, with an exposure of about 3×10^{21} nvt ($E > 1$ MeV) showed no effect of irradiation up to that exposure.⁽¹⁰⁾ The only elevated temperature crack propagation test data available on irradiated material come from three tests on PRTR tubing with exposures of about 2 to 3×10^{21} nvt ($E > 1$ MeV). These tests were run at 20, 100, and 200 °C with

slot lengths of 1 1/2 in. The results were the same as for unirradiated material tested under similar conditions and the crack arrest temperature was found to be between 20 and 100 °C, which is comparable to unirradiated data.⁽¹⁰⁾ There is work being done at Chalk River on crack propagation tests of irradiated material and hydrided irradiated material that should be reported in 1969.

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7.0 RADIATION EFFECTS ON COUPON PROPERTIES

F. R. Shober, R. G. Hoagland, T. T. Claudson,
E. R. Gilbert, R. W. Barker, and H. J. Lowell

Metals and alloys undergo changes when subjected to neutron environments that alter their mechanical and physical properties. The major part of these changes occur as a result of either atomic displacement or transmutation, with the former having the greatest effect on materials irradiated at temperatures below about 932 °F (500 °C). The magnitude of change is a function of total neutron fluence and irradiation temperatures. This section, therefore, discusses property changes in terms of total fluence, irradiation temperature, test temperature, and postirradiation annealing temperatures. Those properties most frequently used for design considerations such as tensile, creep, fatigue, and notch toughness are presented. The effect of irradiation on physical properties is also discussed.

7.1 CREEP AND STRESS RUPTURE

The effect of neutron irradiations on the creep properties of Zircaloy-2 has been studied by several investigators. The more recent work has pertained to in-reactor creep studies rather than postirradiated creep and has been done mainly at two laboratories, Pacific Northwest Laboratory and Chalk River. Some in-reactor biaxial stress tests were made by workers at Oak Ridge National Laboratory. The results of these studies are discussed.

Several significant effects of neutron irradiation on in-reactor creep properties have been noted as the result of recent investigations of Zircaloy-2. The first of these⁽¹⁾ includes in-reactor creep rate data in the 310 to 450 °C temperature range and stress levels from 20,000 to 35,000 psi.

The ratio of reactor-on creep rate to control creep rate varied from 0.42 to 2.2 as shown in Table 7.1. Perhaps the most significant effect observed due to irradiation was an increase in activation energy for creep from 60,000 cal/mole without irradiation to approximately 90,000 cal/mole during neutron irradiation in the temperature range of 325 to 425 °C. These data are shown in Figure 7.1. As a result of this higher temperature dependence of the in-reactor creep rate, the effect of irradiation may either increase or decrease the creep rate depending upon the test temperature relative to the temperature where the crossover occurs on an Arrhenius type plot of the curve for the in-reactor data and the control data. The crossover point is also sensitive to stress and moves to lower temperatures with increasing stress. A plot of strain versus theta parameter, temperature compensated time, is shown in Figure 7.2 for creep rates measured in-reactor during irradiation.

FIGURE 7.1. Activation Energies for In-Reactor Creep During and After Irradiation

TABLE 7.1. Summary of In-Reactor Creep Results

Test Conditions Stress, psi Temperature, °C		Behavior During Reactor Operation	Behavior During Reactor Outages	Ratio of Reactor- on Creep Rate to Control Creep Rate	Reactor-on Creep Rate, per hr	Control Creep Rate, per hr
20,000	350	Always showed lower strain than control; control slowing faster than in-reactor.	No rate changes observed.	2.2 (2000 hr)	7.5×10^{-7}	3.4×10^{-7}
20,000	275	More strain than control; starts faster, but slows down sooner.	Immediate increase in rate observed on shutdown. Magnitude of increase about 1.6 times.	1.5 (500 hr)	2.2×10^{-6}	1.5×10^{-6}
25,000	350	Slightly more strain than control; appears to slow faster than control.	No outage data available.	0.86 (500 hr)	1.9×10^{-6}	2.2×10^{-6}
25,000	367	Considerably more strain than control.	Increase in rate found immediately upon shutdown.	0.75 (400 hr)	9×10^{-6}	1.2×10^{-5}
30,000	310	More strain than control.	Increase in rate occurred. ^(a)	0.50 (500 hr)	5×10^{-7}	1×10^{-6}
30,000	350	More strain than control.	Increase in rate occurred about 25 hr after outage.	2.2 (200 hr)	3.4×10^{-5}	1.5×10^{-5}
35,000	310	More strain than control, but slowing faster.	No outage data available.	1.1 (500 hr)	4.5×10^{-6}	4×10^{-6}
35,000	320	Higher initial strain; with increasing time, this reverses. Control shows increasing rate after 900 hr.	Creep rate increased about 100 hr after shutdown.	0.42 (400 hr)	1×10^{-5}	2.3×10^{-5}

(a) Data for this test are in question.

FIGURE 7.2. Theta Correlation of In-Reactor Data

A similar study was made at Chalk River⁽²⁾ on the influence of neutron irradiation on the creep of Zircaloy-2 at 300 °C and at somewhat lower stress levels. At 300 °C and these lower stress levels, the neutron irradiation was found to cause much faster creep rates than the out-of-reactor control tests. These data are found in Table 7.2.

In-reactor creep work is being conducted in Culcheth, England by B. Watkins. Although not yet published, these results confirm the results reported in Reference 2 in that much faster creep rates were observed during irradiation than in the absence of neutrons.

In-reactor creep tests on pressurized tubes of Zircaloy-2 have been reported.⁽³⁾ As a first approximation, the following equation for the creep rate

$$\text{Creep rate/hr} = 4(10^{-27}) \sigma_t \phi (T-160)$$

can be used for cold-worked Zircaloy-2 in the temperature (T) range from 250 to 300 °C, hoop stress (σ_t) range from 10,000 to 16,000 psi, and with fast-neutron flux (ϕ) n/cm²-sec (E > 1 MeV). These results show that the creep rate is significantly higher due to the neutron flux.

In-reactor creep tests on pressurized tubes of Zircaloy-2 are also being conducted at the Pacific Northwest Laboratory,⁽⁴⁾ results unpublished also show a significant increase in creep rate due to the neutron irradiation.

In-reactor stress rupture tests on pressurized tubes of Zircaloy-2 conducted at temperatures of 371 and 482 °C were reported by Hinkle.⁽⁵⁾ He observed no effect of the neutron irradiation on the time to rupture. His data are shown in Figure 7.3.

TABLE 7.2. Creep of Cold-worked, Stress-relieved Zircaloy-2 at 300 °C;
In-reactor, Flux 5 to 10 x 10¹² n/cm²/sec (E > 1 MeV)

Test No.	Max. Fast Neutron Flux (E > 1 MeV) x 10 ¹²	Test Atmosphere	Source of Specimen	Specimen Diameter, in.	Stress, lb/in. ²	Time in Reactor, hr	Minimum In-Reactor Creep Rate, %/hr	Final Creep Rate of Laboratory Control Tests, %/hr ^(a)
R2	5-8	Air	20% CW rod	0.040	30,000	2500	2 x 10 ⁻⁴	3 x 10 ⁻⁵ (2500)
R4	8	Air	20% CW rod	0.040	20,000	1300	5 x 10 ⁻⁵	3 x 10 ⁻⁶ (1300)
R4	8	Air	20% CW rod	0.040	30,000	1900	2 x 10 ⁻⁴	2.5 x 10 ⁻⁵ (2000)
R6	7-10	Helium	16% CW pr. tube	0.080	20,000	4500	3 x 10 ⁻⁵	2 x 10 ⁻⁶ (4500)
R8 ^(b)	5	Helium	19% CW pr. tube	0.100	15,700	400	3 x 10 ⁻⁵	4 x 10 ⁻⁷ (38,000)
R9 ^(b)	8	Helium	19% CW pr. tube	0.100	11,000	2100	2 x 10 ⁻⁶	-
R9 ^(b)	8	Helium	19% CW pr. tube	0.100	18,000	1400	3 x 10 ⁻⁵	1 x 10 ⁻⁶ (10,000)

(a) Duration of tests in hours is given by the value in brackets.

(b) Specimen tested in the laboratory prior to transfer to the reactor.

FIGURE 7.3. Effect of Neutron Bombardment on Stress-Rupture Strength of Zircaloy-2 at 700 and 900 °F in Helium

A special issue of the Journal of Nuclear Materials is being prepared for publication in the spring of 1968 which will make available more recent data on in-reactor creep of Zircaloy-2.

Stress Relaxation

Stress relaxation tests which are being conducted on Zircaloy-2 both in-reactor and without irradiation are being conducted at Bettis Atomic Power Laboratory and by other laboratories; however, these results have not yet been published in the unclassified literature.

7.2 TENSILE STRENGTH AND DUCTILITY

The tensile properties of unirradiated Zircaloy-2 have been studied by a number of investigations⁽⁶⁻⁸⁾ and provide a basis for comparison with the irradiated tensile properties. The influence of test temperature, anisotropy, and hydrogen content on the tensile and yield strength of annealed Zircaloy-2 are shown in Figure 7.4. The scatter bands reflect the

FIGURE 7.4. The Effect of Test Temperature on Strength of Unirradiated Zircaloy-2. Scatter Bands Indicate Scatter Caused by Additions of H_2 (Random Orientation) of 12, 150, and 500 ppm.

data scatter caused by differences in hydrogen content, with those materials containing greater amounts of hydrogen having values which fall in the upper part of the band. Notice that the strength differences caused by the anisotropy produces two distinct scatter bands.

The data for irradiation effects in Zircaloy-2 fall essentially into two categories, reactor ambient temperature irradiations ($<212^{\circ}\text{F}$) and elevated temperature irradiations ($>212^{\circ}\text{F}$). The latter category as it applies here is about 575°F (300°C). Bement⁽⁹⁾ reports that irradiation hardening in Zircaloy-2 progresses at a markedly different rate at elevated temperatures than at reactor ambient temperatures. It follows then that the lower temperature irradiation effects data are not directly applicable to reactors where service temperatures are $>212^{\circ}\text{F}$. For this reason, emphasis is placed on elevated temperature irradiation data.

Unfortunately, irradiations have not been performed at temperatures greater than approximately 575°F (300°C); neither have mechanical properties data been obtained above 575°F (300°C) for Zircaloy-2 irradiated at elevated temperatures.

Figures 7.5 and 7.6⁽¹⁰⁾ show the effect of irradiation up $8.6 \times 10^{21} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$) on annealed Zircaloy-2 specimens whose longitudinal axes were transversed to the rolling direction of the plate material. The low strain hardenability after irradiation is evident from the similarity of the yield and ultimate strengths shown in Figure 7.5 and the low ductility values shown in Figure 7.6. Uniform and total elongation values are reduced to about 0.5% for irradiation greater than $1.0 \times 10^{21} \text{ n/cm}^2$. Similar changes were noted for 20% cold-rolled Zircaloy-2 after irradiation. These changes are shown in Figures 7.7 and 7.8.⁽¹¹⁾

FIGURE 7.5. The Effect of Exposure on the Strength of Annealed Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

7.11

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FIGURE 7.6. The Effect of Exposure on the Ductility of Annealed Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

FIGURE 7.5. The Effect of Exposure on the Strength of Annealed Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

7.11

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FIGURE 7.6. The Effect of Exposure on the Ductility of Annealed Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

FIGURE 7.9. The Effect of Exposure on the Strength of Cold-Worked Zircaloy-2 (Irradiated at 290 °C in the (All Transverse Specimens)

FIGURE 7.9. The Effect of Exposure on the Strength of Cold-Worked Zircaloy-2 (Irradiated at 290 °C in the (All Transverse Specimens)

FIGURE 7.5. The Effect of Exposure on the Strength of Annealed Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

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FIGURE 7.6. The Effect of Exposure on the Ductility of Annealed Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

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FIGURE 7.7. The Effect of Exposure on the Strength of 20% Cold-Worked Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

7.12

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FIGURE 7.7. The Effect of Exposure on the Strength of 20% Cold-Worked Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

FIGURE 7.8. The Effect of Exposure on the Ductility of 20% Cold-Worked Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

This same general trend is shown for the 40% cold-rolled Zircaloy-2 in Figures 7.9 and 7.10⁽¹¹⁾ where the uniform strain observed is about 1.0%. Similar material taken from the plate longitudinal to the rolling direction is described in Figures 7.11 and 7.12⁽¹¹⁾ and show similar response to irradiation. Uniform strain is never less than 1.0% and is significantly greater than 1.0% for room temperature tests.

Work by Irvin,⁽¹²⁾ using 18, 28, and 30% cold-worked Zircaloy-2, confirmed many earlier observations regarding the similarity between the yield and ultimate strength values and low strain hardening capability after irradiation. These data also indicated that the cold-worked material had slightly better ductility than annealed material after irradiation to 1×10^{20} n/cm². These materials also showed hydrogen pickup during irradiation. The hydrogen content found was 255 ppm after an irradiation of 2.33×10^{21} n/cm².

Kemper and Zimmerman⁽¹³⁾ studied the effect of annealing temperature on the tensile properties of annealed irradiated Zircaloy-2 after irradiation in the MTR at ~40 °C (104 °F). For room temperature tests, recovery of radiation damage at temperatures between 300 and 400 °C (572 and 752 °F) are reported and shown in Figure 7.13. Recovery of radiation damage is manifested by lowering the 0.2% yield stress, ultimate stress, and to some degree the increase in total elongation.

A comparison of Figures 7.5 and Figures 7.7, 7.9, and 7.11⁽¹¹⁾ shows that: while strength after irradiations up to 8.6×10^{21} is independent of prior cold-work, the apparent anisotropy remains, although less pronounced at the 300 °C (572 °F) test temperature. In fact, the results of the anneals at 400 °C (752 °F) (the highest temperature reported) indicate that the material is fully annealed, notwithstanding

FIGURE 7.9. The Effect of Exposure on the Strength of Cold-Worked Zircaloy-2 (Irradiated at 290 °C in the (All Transverse Specimens)

FIGURE 7.10. The Effect of Fluence on the Ductility of 40% Cold-Rolled Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Transverse Specimens)

FIGURE 7.11. The Effect of Exposure on the Strength of 40% Cold-Worked Zircaloy-2 (Irradiated at 290 °C in ETR)
(All Longitudinal Specimens)

FIGURE 7.12. The Effect of Fluence on the Ductility of 40% Cold-Worked Zircaloy-2 (Irradiated at 290 °C in the ETR) (All Longitudinal Specimens)

FIGURE 7.13. The Effect of Annealing Temperature on the Strength and Ductility of Annealed Zircaloy-2, Irradiated to 1.8×10^{20} in MTR at 60 °C ($E > 1$ MeV)

the more mobile defect state of the low temperature irradiation. The effect of annealing temperatures on the properties of irradiated Zircaloy is shown in Figure 7.14. Cold-worked Zircaloy-2 was irradiated at approximately 280 °C (536 °F) and annealed before testing. The results⁽¹⁴⁾ of annealing at 700 °C (1292 °F) appear to provide fully annealed material. These stress values agree well with other investigators' values for annealed Zircaloy-2. The data seem to indicate that cold-work-irradiation recovery can occur at lower temperatures than cold-work recovery alone. No strain data were available.

7.3 TOUGHNESS

Fracture Properties

The plastic flow and fracture properties of zirconium alloys are important to the safe design and operation of pressure bearing components in nuclear reactors. Early work in the field of study was the use of tensile specimens and data derived from slow notch bend and impact specimens.⁽¹⁵⁻¹⁹⁾ More recently, the use of linear elastic fracture mechanics has been made to obtain a more accurate and quantitative approach to the design of pressure bearing members.

Fracture Behavior

Bement, Tobin, and Hoagland⁽²⁰⁾ studied the effects of neutron irradiation on the flow and fracture behavior of Zircaloy-2. In this work tensile, notch tensile, and slow notch bend data were evaluated in terms of the material fracture characteristics. These studies indicated that useful plastic deformation is limited by strain instability and that it is during local necking that multiaxial stress states develop and the mechanisms of ductile fracture operate. Microcracks nucleated at small intermetallic particles after the initiation of necking were found to form pores as a preliminary step to

FIGURE 7.14. The Effect of Annealing Temperature on the Strength of Cold-Swaged Zircaloy-2 Irradiated to 2.7×10^{20} ($E > 500$ eV) at 280 °C

the initiation and propagation of ductile fracture. These ductile fracture processes in Zircaloy-2 are sensitive to testing direction before and after irradiation and crystallographic anisotropy was found to determine the amount of shear and energy absorbed in the fracture process. Neutron irradiation was also found to greatly reduce the total fracture energy due to the loss of strain hardenability and greater confinement of deformation in the necking process.

Notched specimens tested in slow bending exhibited a sharp ductile-to-brittle transition below room temperature and appreciable deformation accompanied cleavage at subtransition temperatures. Irradiation at 60 °C to fluence levels of 3×10^{20} nvt ($E > 1$ MeV) was not found to alter the transition temperature by more than 25 °C. The notch strength at room temperature was found to increase due to irradiation. The results showed that Zircaloy-2 is not greatly embrittled by irradiation alone. The material is strain rate sensitive and under certain combinations of cold-work and irradiation can become notch sensitive.

Fracture Toughness

Fracture toughness measurements on Zircaloy-2 have been obtained using notch bend (NB), single edge notch (SEN), and double cantilever beam (DCB) specimen geometries in the U.S., Canada, and U.K. These values have been obtained at various fluence levels, irradiation temperatures, and material conditions. W. H. Erickson⁽²¹⁾ examined the fracture behavior of Zircaloy-2 and Zr-2.5 wt% Nb using the SEN specimen. For both cold-worked and annealed Zircaloy-2, the fracture toughness at room temperature decreased sharply as the hydrogen concentration went from 0 to 100 ppm. Fracture toughness increased rapidly with temperature over the room temperature to 200 °C range.

Hoagland⁽²²⁾ and Rowe at Pacific Northwest Laboratory found similar results for Zircaloy-2 with 10 to 400 ppm as shown in Figure 7.15 tested in both the transverse and rolling direction using a DCB specimen. Irradiation of this material at 280 °C to neutron fluence levels of about 3×10^{20} nvt ($E > 1$ MeV) also showed a decrease in the fracture toughness parameter, K_{I_C} , with increasing hydrogen content as shown in Figure 7.16. By comparison of Figures 7.15 and 7.16, it may be seen that specimens containing hydrogen levels below 100 ppm showed a substantial decrease in K_{I_C} due to the irradiation. Irradiation did not greatly affect that material containing 400 ppm hydrogen.

The effects of irradiation on the fracture toughness of annealed Zircaloy-2 is shown in Figure 7.17. Double cantilever beam specimens were irradiated at 280 °C to fluence levels of 1×10^{21} nvt ($E > 1$ MeV). A decrease in K_{I_C} with neutron fluence level was found at fluence levels to about 3×10^{20} nvt ($E > 1$ MeV). This effect seems to saturate with only small differences being measured at fluence levels to 1×10^{21} nvt ($E > 1$ MeV). These effects appear to be dependent upon test temperature with the greater effects being prominent at the higher test temperatures.

The effect of cold-work on K_{I_C} has also been determined by Hoagland⁽²²⁾ and is shown in Figure 7.18 for Zircaloy-2 in the 20 and 40% cold-worked condition. For transverse specimens tested at temperatures to -20 °C, the fracture toughness decreases with increasing cold-work. At -20 °C a crossover occurs wherein the 40% cold-worked material has the higher K_{I_C} value. Similar effects are indicated for the specimens tested in the rolling direction. It was also noted that the rolling direction specimen toughness values were lower than those specimens tested in the transverse direction.

Neg. PNL0662399-1

FIGURE 7.15. Fracture Toughness of Hydrogen Charged Zircaloy-2

FIGURE 7.16. Fracture Toughness, K_{Ic} , of Hydrogen Charged Zircaloy-2 Irradiated at 280°C

7.26

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Neg. PNL0661335-10

FIGURE 7.17. Fracture Toughness of Irradiated Zircaloy-2

Neg. PNL0661421-1

FIGURE 7.18. Fracture Toughness of Cold-Worked Zircaloy-2

The combined effects of hydrogen and irradiation were also studied by Erickson and Rechman at Chalk River on annealed Zircaloy-2 with hydrogen concentrations which varied between 400 to 800 ppm. Irradiations at 50 to 100 $^{\circ}\text{C}$ were conducted to fluence levels of 4.3×10^{19} nvt ($E > 1$ MeV). Fracture toughness values were calculated from testing round notch tensile specimens. Little, if any, changes occurred in

fracture toughness of this material due to the irradiation. Additional work in this area is planned at Chalk River using the DCB specimen employed by Hoagland.

G. W. Parry has also studied the effect of hydrogen on the fracture toughness of Zircaloy-2 and Zr-2.5 wt% Nb.⁽²³⁾ This study also showed that hydrogen lowers the fracture toughness at room temperature. This also applied to decreasing the critical crack lengths of zirconium alloys. However, Parry showed that at 300 °C Zircaloy-2 is unaffected by hydrogen concentrations up to 500 ppm.

Critical Crack Length

Subcritical flaws may exist within a pressure bearing structure without disastrous effects on its operating performance. However, these flaws, formed during the fabrication process or nucleated during operation, may grow as a result of fatigue or subcritical crack growth due to the stress. Safe operation of reactor pressure tubes and vessels requires that critical crack lengths be known and detected prior to impending failure.

A. Cowan and K. J. Cowburn⁽²⁴⁾ performed tests on Zircaloy-2 tubes containing artificially-induced through wall thickness longitudinal slits which were pressurized to failure and extensive work has been performed by R. C. Aungst, P. J. Pankaskie, and T. R. Ostrom at Pacific Northwest Laboratory as reported in Sections 5.0 and 6.0.

Correlations between impact energy and failure stress have been made by Pickles and Cowan.⁽²⁵⁾ Impact tests were made on material taken from zirconium alloy pressure tubes on which critical crack length measurements had been made. Tube failure stresses and the corresponding impact energy at the tube burst test temperature were compared. The correlation predicted failure stresses within about 30% of actual values.

Translating these results to irradiated material predicted a critical crack length of 2 1/2 to 3 in. for material irradiated at 300 °C and 16,000 psi with 520 ppm hydrogen. At temperatures below 200 °C, the critical crack length at 16,000 psi will be less than 1 in.

Delayed Failure Analysis

Delayed failure analysis of zirconium alloys is extremely important since it is the nucleation and growth of small, undetectible flaws during static or fatigue loading which may precipitate catastrophic failure. Growth rates of these flaws are sensitive to not only irradiation, stress level, and temperature, but also to material condition and environment. To date, little effort has been expended in this analysis. Initial work conducted by Armour Research Foundation⁽²⁶⁾ using notched and unnotched specimens of zirconium and Zircaloy-2 indicated little effect of hydrogen at the 200 ppm level to delayed failure at room temperature.

7.4 PHYSICAL PROPERTIES

Several studies have been made of various physical properties of zirconium and its alloys after irradiation. Changes observed are limited and should pose no problems in reactor operation.

Electrical resistivity changes during irradiation. The extent of such changes and their recovery by annealing were reviewed by Lustman and Goodwin in the Reactor Handbook.⁽²⁷⁾ Increases in electrical resistivity of 2 to 8% occurred when measured at room temperature, 10 to 30% at -196 °C (-320 °F). Recovery was well advanced at 275 °C (527 °F) and virtually complete at 400 °C (752 °F) after 2 to 10 hr.

The in-reactor measurement of electrical resistivity of zirconium in the annealed and 90% cold-worked condition have

been reported by Provdyuk and Ivanov.⁽²⁸⁾ Resistivity increases were 7.5% at 2.9×10^{19} n/cm² when the zirconium was in the annealed state; about 4.5% when cold-worked. The change in the annealed sample corresponded to that introduced by 80 to 90% cold-work. In-reactor resistivity measurements revealed that the slope at the initial increase in electrical resistivity versus neutron fluence was comparable to the long-term slope. This is unlike molybdenum and tungsten where the initial slope is much greater than the slope at higher fluence levels. Recovery experiments after irradiation revealed that some decrease in resistivity occurs below 100 °C (212 °F) with uniform rate of resistivity decrease occurring to 340 to 400 °C (644 to 752 °F) where the final damage is removed. These curves apply to recovery of electrical resistivity in both the annealed and cold-worked irradiated states and the cold-worked unirradiated states. Apparently, the defects generated during irradiation are rather complex, as indicated by the uniform recovery curve and high temperature for final recovery or irradiation-induced defects.

Experiments have been made to determine the effect of irradiation on dimensions and densities of zirconium and its alloys. Changes observed have been generally within the limits of experimental error for techniques used to measure these changes. Similar results have been obtained for changes in elastic moduli, where again most differences between irradiated and unirradiated values could be attributed to experimental error.

Very little can be said concerning irradiation-induced changes in microstructure of X-ray diffraction patterns. Optical and electron microscopy studies reveal no differences. X-ray lattice parameter and line breadth measurements are reported by Howe⁽²⁹⁾ for zirconium. Irradiation to a neutron fluence of 1.4×10^{20} n/cm² ($E > 500$ eV) at 50 °C (122 °F)

increased the "a" parameter 0.019%. There was no change in the "c" parameter, no line broadening, and no density change. Based on indirect observations of fuel elements and the creep process, there is evidence of pronounced increases in diffusion during irradiation.

A critical factor influencing the properties of zirconium is its hexagonal-close-packed crystallographic structure; its properties are particularly sensitive to fabrication history. Because of this, it is often difficult to interpret irradiation effects on alloys of an unknown fabrication history.

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8.0 TRANSITION JOINTS BETWEEN ZIRCALOY-2 AND PRIMARY COOLING SYSTEM

L. A. James

8.1 INTRODUCTION

In the design of nuclear reactors, the use of Zircaloy-2 pressure tubes is economically justified only in the areas of high neutron flux. The transition between Zircaloy-2 and the primary cooling system material is therefore made as close to the active reactor core as practical. The joint between the two materials must be leak-free, with a high degree of reliability to meet economic and safety requirements. The design of this joint or transition to these criteria is difficult because Zircaloy-2 and normal piping materials have large differences in coefficients of thermal expansion which result in large thermal stresses of the mated materials during reactor operation. Many of the attempts at gasketed joints have had unacceptably high leakage rates, and many of the attempts at achieving a metallurgical bond have been plagued by the formation of brittle intermetallic compounds at the joint interface. This section will review some successful, as well as unsuccessful transition joint designs.

8.2 JOINT EXPERIENCE

Gasket Joints

A number of gasketed closures have been evaluated in the past.⁽¹⁻³⁾ These include "O" rings, delta rings, "banana" rings, Bridgeman, "dome" seals, Greylocs, Conoseals, delta seals and spiral-wound asbestos-stainless steel gaskets. Most of the various systems were found to seal effectively; however, the majority of them require high quality mating surfaces which could be damaged easily by maintenance practices. The spiral-wound asbestos-steel gaskets have been used successfully in the

PRTR reactor at Hanford⁽³⁾ and in the NRU reactor at Chalk River.⁽⁴⁾ These gaskets do not require the high quality surfaces necessary for some of the other designs, and they are easy to maintain and replace. Some evidence of stress corrosion cracking was noted in the stainless steel winding of the gaskets used at Hanford.⁽³⁾ The Marmon Conoseal has been used successfully in the CVTR.⁽²⁾ The leakage design goal for this system was 1 lb of heavy water/year/joint.

Fusion-Welded Joints

In general, attempts to fusion weld Zircaloy-2 to steel have been unsuccessful due to the formation of brittle intermetallics at the joint interface. These result in joints of low strength and ductility.⁽⁵⁾ A recent development at the Atomic Energy Establishment at Harwell⁽⁶⁾ achieves a weld between the two materials by first TIG-welding niobium to the Zircaloy-2, and then brazing the niobium to stainless steel. Little information is given about this process, and it is not known if any such transition sections have been installed in a reactor. A combination of gaskets and fusion welds have been used successfully at Chalk River⁽⁴⁾ to make the transition from Zircaloy-2 to stainless steel. The Zircaloy-2 pressure tube ($\alpha = 3.40 \times 10^{-6}$) is joined to a section of 43% nickel-iron ($\alpha = 3.59 \times 10^{-6}$) with a flanged joint employing spirally-wound asbestos-iron gaskets. The small difference in alpha allows a standard flanged joint to be used. The 43 Ni-Fe is then fusion-welded to 50 Ni-Fe ($\alpha = 5.55 \times 10^{-6}$), which in turn is fusion welded to 60 Ni-Fe ($\alpha = 6.86 \times 10^{-6}$). The 60 Ni-Fe is then fusion-welded to Type 347 SS ($\alpha = 9.43 \times 10^{-6}$) using Inco-weld "A" ($\alpha = 7.35 \times 10^{-6}$). This scheme avoids the problem of brittle intermetallics at the Zircaloy steel interface, and the difference in thermal expansion between any two sections

is not great enough to cause problems. Difficulty was experienced, however, in achieving satisfactory welds between the various sections of Ni-Fe alloy.⁽⁷⁾

Brazed Joints

Since zirconium tends to form brittle intermetallic compounds with nearly all elements at elevated temperatures (iron, chromium, nickel, etc.), attempts to achieve a brazed transition joint have had the same general results as the welded joints. Fairly high strengths are possible, but the joints possess low ductility.^(5,8)

Diffusion Bonding

Diffusion bonding has been somewhat more successful than the welding or brazing process.⁽⁸⁻¹⁰⁾ The lower temperatures result in small zones of brittle intermetallics at the interface. It has been found⁽¹⁰⁾ possible to produce thin diffusion zone (less than 5 μ) bonds with acceptable mechanical properties. The thickness of the diffusion zone may be controlled to a certain degree by varying the "incubation period" for diffusion. Successful bonds were also reported⁽⁹⁾ where the joints withstood 200 thermal cycles (100 to 600 °F) and 100 pressure cycles (100 to 3500 psi) without failure. Hydraulic bursting pressures of 13,200 to 17,200 psi (hoop stresses of 40 to 52 ksi, respectively) were required to cause failure, and the failure generally occurred by the Zircaloy-2 splitting longitudinally and peeling away from the joint.

A form of diffusion bonding may be accomplished by the process of tandem extrusion.⁽¹¹⁾ A scarf joint between the Zircaloy-2 and the stainless steel is extended with an area reduction ratio of 6:1 at temperatures from 1500 to 1650 °F. The lower exposure time for the dissimilar metals at elevated temperatures should give the process some advantage over a more conventional diffusion bond. Also, the shearing of the

surfaces past each other should break up oxides and other films. However, scatter in the test data for mechanical properties of the joints produced⁽¹¹⁾ suggest that either brittle intermetallics were formed, or that the process was not perfected at that time. Satisfactory tandem extruded joints were evaluated at Hanford.⁽¹²⁾ During burst testing, failures occurred in the parent materials, indicating a sound joint. Extruded joints have also performed satisfactorily after exposure to radiation.^(13,14)

Rolled Joints

Transition joints consisting of a Zircaloy-2 tube rolled into a steel nozzle have been successfully used in various Canadian reactors, as well as the N-Reactor at Hanford.⁽¹⁵⁾ The joints thus formed are "mechanical" rather than metallurgical, because the components are forced together in an interference fit and little or no atomic bonding is accomplished. The steel nozzle was grooved and the Zircaloy-2 tube was rolled into these grooves. This provides better sealing characteristics and also provides axial load carrying capability. The joints met all structural requirements and properly-formed joints did not leak. An attempt was made to achieve metallurgical bonding in rolled joints.⁽¹⁶⁾ It was found that the metallurgical bond was feasible with roll forming, but it was found that the use of a soft-metal inlay gave added assurance of leak-tightness. As of October, 1967, there have been no leaks observed in the rolled joints installed in the N-Reactor at Hanford, and performance has been satisfactory in every way.⁽¹²⁾ Examination of joints in service has indicated no significant stress relaxation of the interference fit between the dissimilar materials. Officials at Chalk River, Ontario, expressed satisfaction with the rolled joints in the NRU, NRX and NPD reactors, and stated that rolled joints are contemplated in all future reactor designs.⁽⁷⁾

Explosive Bonding

The process that appears to offer the most promise for future applications is explosive bonding. The bonding may be accomplished at room temperature, and the metallurgical bond that results is not plagued with the brittle intermetallics that are associated with the heat-dependent processes. The process is essentially a nonplanar interface collision resulting from the progressive detonation of an explosive charge. At the point of collision, an effect known as "jetting" causes the surface films to be "squirted" out ahead of the advancing collision point. This results in a film-free interface and essentially diffusionless metallurgical bond is created. Explosive bonding has been successfully used to join Zircaloy-2 to stainless steel, Inconel, aluminum and several other reactor materials.⁽¹⁶⁻²⁰⁾ Surface finish requirements are not critical (1000 μ in rms or less), and the surfaces need only be wire brushed and degreased prior to welding. The advantages of this process might be lessened if operating temperatures were high enough to allow the formation of brittle intermetallics over a long period of time. The effect of irradiation on explosively welded joints, as well as tandem extruded joints, was evaluated,⁽¹³⁾ and it was determined that the structural integrity of the joints was not impaired. The Canadians have combined the explosive bonding and roll-forming processes by first achieving a partial bond by explosive means and then by finishing the joint by roll-forming.⁽⁷⁾

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9.0 FUEL SUPPORT-PROCESS TUBE INTERACTION

D. P. O'Keefe

In the operation of the PRTR and the N-Reactor, problem areas which have commanded extensive technical attention are associated with the requirement for positioning fuel within the coolant stream in a manner that permits easy charge-discharge. This requirement is most simply met by letting the fuels be loosely positioned by supports of the proper dimension to maintain the coolant annulus. Complications arise from the fact that the fuel element, coolant, and pressure tube are a dynamic system during reactor operation and during charge-discharge, and as such include the parameters of dynamic loads and mechanical wear. The pressure tubes and the fuel element support hardware in both reactors are constructed of Zircaloy-2, which under some of the operating conditions of the PRTR has demonstrated its susceptibility to fretting corrosion. In the N-Reactor a complex series of events including the wear of sliding parts in combination with fretting corrosion have combined to present problems for solution by the reactor operator and/or fuel designer.

In the PRTR each of the 85 vertical fuel channels contains one fuel element assembly consisting of a 19 rod cluster about 8 ft long. The rods are Zircaloy-2 clad Al-Pu alloy or ceramic, and weigh a total of 70 or 140 lb, respectively, per assembly. The 19 rods are held in the desired arrangement in the cluster by Zircaloy-2 end brackets at the top and bottom. The Al-Pu and most of the ceramic fuel clusters are bound together with circumferential bands. Some of the ceramic fuels are autoclaved before assembly and bundled together with 0.072 in. wire in a circumferential tie or a spiral wrap. The fuel element assembly is positioned vertically in the pressure tube by attachment to a hanger rod which is suspended from the outlet nozzle at the top of the tube. The heavy water coolant flows

upward at a maximum rate of 123 gal/min with outlet temperature and pressure of 277 °C and 1080 psig. The pressure tubes are Zircaloy-2. They are autoclaved to provide a very hard, essentially chemically inert zirconium dioxide film.

At the time Zircaloy-2 was selected as the material for the pressure tubes, there was very little operating data on the performance of the alloy in service conditions similar to the PRTR. Therefore, a surveillance program was initiated which included periodic, in-reactor visual examinations of the internal surface of the tubes. This was accomplished with a television camera-borescope arrangement which permitted observation of the tube surface and measurement with a dial indicator of discontinuities in the surface. The results of this surveillance program over 3 yr is summarized by Winegardner⁽¹⁾ in a detailed account of the process tube-fuel support interaction experience in the PRTR which is the major source of information presented here on this subject.

After only 2 weeks of operation, vertical fretting marks were found on the walls of the first seven tubes examined. Subsequent examination of 83 tubes showed spiral marks where fuel bundle wraps or individual rod wire wraps contacted the tube walls. Examination of selected tubes showed that with increased reactor operating time, the number of marks increased and the depth of penetration of the tube wall became greater.

After a year of operation, all of the pressure tubes had been examined and 1600 fretting marks were found. Most of the marks were less than 0.006 in. deep while 2.2% of the marks were deeper than 0.010 in. The maximum penetration of 0.026 in. associated with a bundle wire wrap, occurred in approximately 2 months of operating time. Approximately twice as many fretting marks were found at the bottom end bracket location as were found at the upper end bracket location for tubes containing the lighter Al-Pu fuel elements. In addition, penetration was

deeper at the bottom end bracket location. For tubes containing the heavier UO_2 fuel elements, approximately the same number of marks was found at the top end bracket location.

It was felt that the severity of attack in the reactor could be decreased by increasing the surface contact area, thereby reducing the stress intensity at the area of contact. An in-reactor test with an element with wider contact pads to center the element in a new tube indicated that this measure effectively reduced the rate of wear. Subsequently, most of the fuel elements in the PRTR were equipped with centering feet which provided a contact area 1/4 in. by 1/2 in. long. Provision was made for equipping irradiated fuels with snap-on centering feet.⁽²⁾ This measure reduced the severity of the fretting corrosion attack. New areas resulting from fretting continued to appear, but they were nearly all quite shallow (< 0.001 in.). By 1966 only six process tubes exhibited fret marks 0.010 in. or more in depth.⁽³⁾

Although the wider contact pads appeared to be effective in reducing the attack between fuel supports and pressure tubes, the situation was not entirely that simple as indicated by the fact that several tubes were left charged with elements having narrow supports (1/16 in. wide) and the rate of fretting in these tubes also decreased. In looking for possible changes in the reactor operating environment which might have influenced the process, it has been suggested that replacement of a flow straightener on a lower ring header may have contributed to the change in fretting rate. The flow straightener served the purpose of reducing turbulence induced by the centrifugal pumps. The initial installation failed during early PRTR operation. This was replaced with a new installation having a different design at about the time that tests of fuel elements with increased contact areas were starting. It was felt that the failed flow straighteners could have been the source of

increased vibration in several of the pressure tubes. Variation in flow rate that may have resulted from the new installation was thought to be unimportant, because intentional flow increases by reorificing did not influence the fretting rate. Four tubes with a 7% flow increase did not show any change, even with fuels having 1/16 in. wide feet.

One design change which eliminated the severe fretting associated with the spiral wire bundle wraps on the oxide fuels was the use of a circumferential strip band wrap. The spiral wire bundle wrap did not sufficiently contain the rod bundle and this increased the possibility for contact between the wire wrap and the pressure tube. Replacement with circumferential wraps maintained the fuel cluster in a more restrained assembly and restricted the contact points to the end bracket pads.

To summarize the PRTR experience, it may be stated that in 250 °C and 1000 psig water Zircaloy-2 may be subject to fretting corrosion wherever mating surfaces have any appreciable relative motion. The severity of the attack in the PRTR case was influenced by changing the area of contact between fuel supports and pressure tube, and also by changing reactor operating characteristics which influenced the driving force between the fuel and tubes. The most recent conclusion as the result of several years of reactor operation is: "The PRTR process tube surveillance program has shown that a variety of fuel element weights, lengths and pad design can be irradiated in the PRTR without undue damage to the process tubes if adequate fuel element designs are utilized and normal PRTR operating conditions are applied."⁽³⁾

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10.0 OPERATIONAL STANDARDS AND SURVEILLANCE

P. J. Pankaskie

10.1 INTRODUCTION

Within the U.S., there are at least three water cooled reactors, operating at power producing temperatures, which use zirconium alloy pressure tubes in the core sections of the primary coolant circuit. In Canada, there are several similar reactors now operating or in advanced stages of construction. In the U.K., there is a steam generating reactor which also uses some Zircaloy-2 pressure tubes in the primary coolant system.

In these reactors, the number of pressure tubes in a single reactor range from eighty-five to nearly one thousand. In the construction of these nuclear reactors, both the type and scale of application of Zircaloy-2 pressure tubes were unprecedented with prior experience limited to special test loops in several operating reactors. The absence of a significant history of operating experience in both a nuclear and non-nuclear environment provided the incentive for undertaking pressure tube surveillance programs at several of these reactors. In most instances, the surveillance programs consist of periodic in-reactor inspections and destructive testing and evaluation of pressure tubes removed at specific increments of service.

This section will describe briefly the tube related operational standards and the surveillance programs in effect at the various reactors and the instrumentation used for in situ pressure tube inspections.

10.2 OPERATING STANDARDS

Operating standards which pertain to, or which limit, reactor operation because of the presence of zirconium alloy

pressure tubes are difficult to obtain for individual reactors. The Canadian experimental (NRX, NRU, NPD) and power reactors (CANDU-type) apparently have no written standards for reactor operation which relate directly to the pressure tubes;⁽¹⁾ nor do any Canadian or Hanford pressure tube reactors (PRTR and N-Reactor) have written specifications for items specifically investigated during tube inspections.^(1,2)

The PRTR and N-Reactor do have rather specific inspection programs which state the frequency of examination, normal sampling procedures to select tubes for examination, and criteria which require specific tubes to be examined because of non-normal operation history.⁽³⁻⁶⁾ This would include tubes which have experienced a stuck fuel element, a fuel rupture, a malfunction by temperature or flow trip, or a situation where a reasonable basis exists for suspecting that something detrimental may have happened to the tube.⁽⁵⁾

During inspection, the tubes are visually examined, with borescopes, for unusual items such as corrosion, fretting, blisters, or scratches and are checked for either inside diameter or circumferential dimensional changes. If anything unusual does appear, a technical analysis is made and a decision is made as to how to proceed.

There are no adopted criteria for tube removal. Such criteria have been written for the PRTR tubes, but have not yet been formally accepted.⁽⁷⁾

The PRTR⁽⁸⁾ and N-Reactor have operating standards, including startup and shutdown procedures, which relate to the pressure tubes. These standards are written to allow each tube situation to be considered individually. For example, the N-Reactor maintenance standards state "any incidence or occurrence

which may lead to the consideration of a pressure tube for replacement shall be referred to plant Engineering and Research and Engineering."

In general, operating pressure and temperature standards, coolant chemistry, and startup and shutdown procedures are limited more by the out-of-reactor primary piping and valves than by the Zircaloy-2 tubes.

10.3 IN-REACTOR SURVEILLANCE PROGRAMS

PRTR

There are 85 Zircaloy-2 pressure tubes in the PRTR core section of the primary coolant circuit. During normal reactor operation, most tubes contain fuel elements. That part of the pressure tube into which the fuel element is placed is nominally 3.25 in. in diameter on the inside. One end tapers to a nominal 1 1/2 in. ID. Actual dimensions of the pressure tube are shown in Figure 4.1, Section 4.2. The tubes are vertical and each is surrounded by a thermally insulating helium gas annulus which is enclosed in an aluminum tube. The arrangement of the pressure tube in the reactor core is shown in Figure 10.1. Fuel elements are charged and discharged from the pressure tube through the nozzle and nozzle cap assembly at the top end of the tube.

The design of the Zircaloy-2 pressure tubes was based on an allowable creep deformation of 1%. This deformation limit was selected because little stress rupture data were available; the strain limit was considered to be conservative in relation to the reduction in ductility due to radiation damage and this magnitude of deformation could be measured in situ by measuring the inside diameters of the Zircaloy-2 pressure tubes. Hence, this design criterion formed much of the basis for periodic inspections of pressure tubes in situ.

Neg. C3582-1

FIGURE 10.1. PRTR Pressure Tube Assembly

Hydraulics investigation, with a prototypic pressure tube and fuel element, showed that minor fretting corrosion could occur and also that some ID surface scratching, from the insertion and removal of fuel elements, could be expected in the PRTR. Based on these observations, some means for visual examinations of the ID surface of the pressure tube was considered essential.

The Zircaloy-2 pressure tube in the PRTR is essentially "hung" from the top flange end (see Figure 10.1). Lateral displacement at the lower end is restricted by the shield plug and inlet jumper. Except for these end restraints, there were no other lateral restraints and bowing of either the aluminum shroud of Zircaloy-2 pressure tubes could occur. If there were sufficient bowing of either tube (less than 0.3 in. lateral displacement), the aluminum and Zircaloy-2 tubes would contact. Heat transfer considerations indicated that boiling at the moderator side of the aluminum tube could occur. Boiling at the OD surface of the aluminum shroud tube could promote intergranular corrosion and ultimately heavy water leakage from the moderator. Such an incident posed no nuclear hazard but from an operational and economic viewpoint, it was most undesirable. Preliminary investigations suggested that eddy current techniques could provide an instrument which would measure the position of the Zircaloy-2 pressure tube within the aluminum shroud tube.

Based on all of these requirements, the examinations and measurements to be made on the pressure tubes in-situ were as follows:

- Visually examine the ID surface of the pressure tube for evidence of mechanical damage, localized fretting corrosion film buildup.

- Measure the inside diameter and/or wall thickness to determine uniform creep deformation and locate areas of local wall thinning.
- Measure the displacement of the axis of the Zircaloy-2 pressure tube from the axis of the aluminum shroud tube as an indication of the eccentricity of the gas annulus between the ID of the aluminum shroud tube and the OD of the Zircaloy-2 pressure tube.
- Measure the zirconium-95 isotope radioactivity in the effluent primary system cooling water as an indicator of unusual corrosion conditions.
- Detect general and/or local hydriding of the Zircaloy-2 pressure tubes.

The initial in-reactor monitoring schedule was based on inspecting one-half of the pressure tubes twice within the initial year of operation. Under this schedule, two tubes from each of three alternate sextants were inspected at the end of each 10-day operating period with repeat inspection after three and before seven consecutive operating periods. Any tube in which a fuel element rupture occurred would be visually inspected prior to reactor startup.

The overall monitoring program also provided for the evaluation of certain pressure tubes by destructive testing after incremental periods of PRTR services. The initial removal and destructive testing and evaluation schedule provided for the removal of two tubes each year for about the first 2 years of operation. This removal rate was subsequently reduced to one per year and now stands at 40 to 200 MW of incremental service exposure, usually 100 MW.

Of the five inspections proposed, only three (visual and ID and gas annulus measurements) were undertaken under the initial schedule and concurrent with PRTR startup. All inspection instruments were incorporated into a single probe which could

be inserted into the process tube in a manner similar to fuel element charge-discharge. Shortly after PRTR startup, visual inspections were augmented with the addition of a remotely operable dial indicator for the purpose of measuring the depth of scratches and fretting corrosion marks.

Early in 1962, inspections indicated a trend of increasing occurrences and severity of local fretting corrosion at points of contact between fuel elements and the ID surface of the pressure tube.⁽⁹⁾ A full reactor inspection was made in June of 1962 and several of the more severely fretted tubes were removed for destructive testing and evaluation.⁽¹⁰⁾ Subsequent to this, the inspection schedule was changed to provide for the inspection of each tube at least once each year. At the same time, experiments were done which showed that the severity of fretting corrosion could be substantially reduced by enlarging the area of each point of contact between the fuel element and the ID surface of the Zircaloy-2 pressure tube.

The schedule of inspecting each tube at least each year continued for almost two more years. During this period very little severe fretting attack occurred although the rate of incidence probably did not change. Evidently, the increase in surface area of the fuel elements supports did effectively reduce the severity of fretting.⁽¹¹⁾ As a consequence, the inspection schedule was revised to provide for inspection of 80% of the pressure tubes each year and all process tubes are examined in each 2 year period.⁽¹²⁾ Current inspection results suggest that further decreases in the required inspections could be considered.⁽¹³⁾

In conjunction with the pressure tube inspections, an effort was made to monitor the radioactivity of the effluent of pressure tubes for the zirconium concentration as a method

for detecting significant fretting corrosion.⁽¹⁴⁾ Water samples that were obtained are concentrated and analyzed by emission spectroscopy. This method is sensitive to zirconium concentration as low as 0.1 ppb. Such analyses lose D₂O but they were generally found to correlate with other general observations of fretting corrosion activity in suggesting that in the PRTR the fretting attack was intermittent rather than continuous. With the rapid bulk mixing of D₂O coolant, this technique is incapable of pinpointing pressure tube by pressure tube where fretting was and was not occurring. Other schemes considered were:

- Continuous bypass of a small portion of the coolant stream into a continuous analyzer where the zirconium content is determined by colorimetric procedure. As yet this scheme is unreliable in concentration ranges of 0 to 10 ppb and depends on the physical state of the corrosion product (e.g. ionic, colloidal, particulate, etc.)
- Continuous bypass of a small portion of the coolant stream into a radioactivity analyzer to detect the concentration of the zirconium-95 isotope.

Further work needs to be done on both schemes before the feasibility of "on-line monitoring" can be determined.

Inspection of the pressure tubes in the PRTR on the scale undertaken was fully as unprecedented as was the use of Zircaloy-2 pressure tubes in the core section of the primary coolant circuit. For nearly the first 4 years of the in-reactor inspection program, all efforts were done by research and development personnel not directly involved in the operation of the reactor. For the past several years, all inspection efforts were done by reactor operating personnel with some consultation from R&D personnel.

On the whole, the in-reactor monitoring effort proved worthwhile in the PRTR. Visual examinations proved to be an

invaluable tool in detecting the extent and magnitude of the fretting corrosion problem and also proved to be quite useful in the effort undertaken to alleviate the problem. Visual examinations detected blister type defects on the ID surfaces of three pressure tubes which had gone undetected through thorough preinstallation inspections. Visual examinations were indispensable in handling the fuel element-pressure tube failure in 1965.⁽¹⁵⁾

About 3 years after the start of the reactor monitoring program, both ID and gas gap measurements were discontinued. Inside diameter measurements disclosed only a little creep deformation. Independent R&D programs on Zircaloy-2 tubing showed that little or no creep deformation was to be expected at stress and temperature conditions present in the PRTR. Hence, there was no further need for ID measurements. With regard to gas gap measurements, little or no permanent warpage of either the pressure or shroud tube was observed with the exception of one or two process channels. One must recall, however, that measurements were obtained with the reactor in a shutdown condition and at or near room temperature. Hence, these measurements may not truly represent conditions when the reactor is operating. In consideration of these two factors, routine gas gap measurements were discontinued.

In the matter of hydride detection, a really suitable instrument has not yet been developed. Examinations of pressure tubes removed from service have disclosed some local hydriding at areas where fretting corruptions occurred. Destructive testing, however, shows that the extent of hydriding so far does not appear to have adversely affected mechanical properties of the tubing to any significant degree. For the present there are no plans to include hydride detection in the routine in-reactor inspection schedules.

As initially conceived, the in-reactor inspection program was rather ambitious. There was no need for ID measurements on a statistically large number of pressure tubes. Measurements on several tubes would have been adequate. Gas gap measurements were not entirely adequate in that they may not truly represent actual operating conditions. They did, however, provide a degree of assurance. As yet there is no really satisfactory substitute other than the installation of some contrivance that physically precludes contact between the pressure and shroud tubes.

One of the greatest difficulties was combining all inspection and measuring instrumentation into a single inspection probe. Separating the instrumentation for each of the inspection functions would have greatly simplified construction and subsequent maintainance of equipment. Separation would also allow much greater freedom in scheduling inspections and make it easier to incorporate changes that experience showed to be useful and desirable.

Visual examinations were made with a borescope coupled to a standard closed circuit television camera. This system produces a two dimensional "view" of the inner surface of the pressure tube. With experienced personnel, this system provides a quick scan of the inner surface of the tube. When the tube surface is not heavily filmed, fretting corrosion "marks" can be readily seen and identified with appropriate lighting. Unidentifiable objects on the inner surface of the tube could be viewed directly through the borescope by means of a "right angle eyepiece." For inexperienced operators, the television system is less than adequate. With the television system there is no color to aid in "scene" identification. Optical resolution and clarity is less than with a borescope alone. A television camera responds most readily to the brightest part of the scene in view, hence dark parts of the scene may not be seen at all.

For an inexperienced operator a standard borescope also presents some significant difficulties. Here again, the view is not truly three dimensional. Axial distances invariably appear to be significantly less than actual. Under certain conditions the image may be reversed and/or inverted thus making it difficult to establish a frame of reference. Despite these difficulties, visual inspections are still considered to be the most useful of currently available inspections methods.

N-Reactor

The N-Reactor utilized about 1000 Zircaloy-2 pressure tubes in the core section of the primary coolant circuit. The pressure tubes are placed horizontally within a stack of graphite and are approximately 50 ft in length. Fuel elements are inserted from one end of the pressure tube and removed from the opposite end. The tube is uniformly 2.70 in. ID with a wall thickness of 0.270 in.

As was the case with the PRTR, N-Reactor pressure tubes were designed primarily on the basis of allowable creep deformation. Based on the few years of prior PRTR experience and additional laboratory data for both irradiated and nonirradiated Zircaloy-2, the allowable creep deformation selected was 1%.

Based to a large extent on PRTR in-reactor monitoring experience, the N-Reactor monitoring effort was designed to provide for:⁽¹⁶⁾

- Visual inspection of the ID surface of the pressure tubes at periodic intervals. A minimum of 2% and up to as high as 5% of the tubes are to be routinely inspected per year under normal reactor operating conditions. Inspections of specific tubes are made whenever a fuel element rupture occurs near the pressure tube wall, or if there were stuck

fuel elements in the tube, or if prolonged hot coolant or severe local power cycling was found to occur in the specific pressure tube.

- Inside diameter measurements of about 2% of the pressure tubes each year under normal reactor operation. Based on visual examinations, ID measurements may also be obtained for specific pressure tubes subjected to any of the aforementioned abnormal operating conditions.
- Routine measurements with eddy current instrumentation for the detection of severe general or localized hydriding (e.g., 1000 ppm H_2).

Under normal reactor operation, the monitoring efforts also requires the removal of two pressure tubes each year for postirradiation examination. A specific tube will be candidate for removal if:

- A fuel element rupture occurs in contact with the ID surface of pressure tube.
- Severe physical damage occurs so as to produce a gouge, scratch, etc., which is 0.025 in. or greater in depth.
- Evidence of severe (1000 ppm or more H_2) local hydriding.
- There is general or local deformation in excess of 2%.

Current in-reactor inspections have been limited to visual examinations. The visual examinations are made by both closed circuit television and direct viewing with an optical borescope. To date, inspections disclosed nominal amounts of fretting corrosion and scratching from fuel element insertion and removal.

Thus far, two pressure tubes have been removed for post-irradiation testing and evaluation. This work is classified and, therefore, it is not reported here.

AECL Reactors

All of the Canadian power reactors utilize pressure tubes. For the reactors already constructed and operating, all used

Zircaloy-2 pressure tubes. Reactors in the design stage and under construction may use Zircaloy-2 or Zr-2.5 wt% Nb alloy pressure tubes. In the NRX and NRU, the pressure tubes are in vertical position. In most, if not all of the power reactors, the pressure tubes will be in a horizontal position.

The current in-reactor surveillance program is limited to the test loops in NRX and NRU.⁽¹⁾ Current schedules call for inspection of the 3-loop tubes in NRU every 2 to 4 months. The single loop tube in NRX is inspected every 4 months. In the NPD, one tube was inspected in 1963,^(1,17) and the same tube was inspected again in 1965. It is anticipated that the same tube and perhaps more will be inspected about every 2 years. These inspections are done by technical personnel at Chalk River Nuclear Laboratory (CRNL). For the power reactors, the utility operator is expected to be responsible for whatever surveillance program he feels to be necessary. CRNL would supply no more than technical consultation.

One of the primary reasons CRNL undertook the in-reactor surveillance effort was to measure in-reactor creep on pressure tubes. Design stresses for the pressure tubes in NPD and other Canadian reactors are appreciably higher than was the case with PRTR and N-Reactor in the U.S. With precision ID measuring equipment, neutron flux effect on in-reactor creep has been observed. As a consequence, the AECL program has been modified to provide for ID measurement on pressure tubes in NRU and NRX test loops which operate at hoop stresses ranging from about 16,000 to 25,000 psi.

All factors considered, the current AECL in-reactor surveillance program is more orientated toward the acquisition of research data than as a reactor operating aid.

Postirradiation evaluation of pressure tubes removed from operating reactors has not yet gotten underway. Two tubes, however, have been removed from the NPD.

The inspections which are done in the NRX and NRU loops are:

- Surface defect and roughness gaging. Instrumentation provides a measure of scratch depth, pits and other surface defects.
- Visual examination of the ID surface by direct viewing through a borescope. Inasmuch as substantial time (several hours) is required, borescoping is not done unless surface defect gaging indicates a need for visual examination.
- ID measurements to detect creep deformation.
- Wall thickness measurements. As yet no wall thickness measurements have been obtained from pressure tubes in-reactor.
- Measurement of bowing of the pressure tube by optical sighting of a target as it is moved along the length of the pressure tube.

The surveillance program has, so far, disclosed little or no fretting corrosion or scratching of the inner surface of the pressure tubes in NRX, NRU, and NPD. Diameter gaging discloses small ($\sim 0.1\%$) but discrete increases in pressure tube ID in the in-core section of the pressure tube. Plotted curves of the increase in pressure tube ID along the length of the tube through the reactor core show the same shape as do curves of neutron flux intensity through the reactor core. Bow measurements indicate that there are no significant problems with bowing.

10.4 EQUIPMENT

Instrumentation for operational surveillance of pressure tube type reactors has primarily been designed for use in the pressure tubes in situ and when the reactor is in shutdown status. As yet there is little "on-line" instrumentation for monitoring behavior during reactor operation. The only known instance of "on-line" monitoring is the measurement of vibration of pressure tubes in the PRTR by means of standard commercial accelerometers attached to the inlet jumpers of the primary coolant circuit.

The inspections of pressure tubes made during a normal or special reactor outage generally consisted of:

- Visual inspection of the ID surface and/or measurement of surface defects, fretting corrosion, scratch depth and the like.
- Dimensional measurements, ID, wall thickness, and gas gap (PRTR only).
- Hydride detection, local and/or general.

Within the U.S., the main inspection method was and still is, visual inspection and surface defect measurements. In Canada, the greatest emphasis is placed on ID measurements and surface defect measurements; relatively little emphasis is currently placed on visual inspection. These differences probably reflect the differences in operating experience at the various reactors.

Visual Inspection Equipment

The system initially designed for the PRTR used an optical borescope through which the ID surface of the tube could be viewed directly or remotely by means of a closed circuit TV camera and monitor.⁽¹⁸⁾ The closed circuit television (CCTV) system provides a means for rapidly scanning the ID surface of the pressure tube and further provides for simultaneous viewing

by two or more operators. The view presented on the CCTV monitor has no color. Lighting of the surface of the tube under inspection must be carefully done to avoid glare since the CCTV camera responds to the most intense light emanating from the object in view. Hence, under certain conditions, a critical part of the view may be "washed" out because it is too bright or too dark in comparison with some other part of the view. In short, a TV camera falls far short of the ability of the eye to accomodate rather wide differences in light intensity within an area in view. To make the adjustments in lighting and CCTV adjustments requires considerable skill on the part of the operator. Optical resolution of a standard commercial CCTV camera is appreciably less than for most optical borescopes.

Both the CCTV camera and the more or less standard monocular borescope present essentially a "two dimensional" view of the area within view of the borescope's objective lens. Distances parallel to the axis of the objective lens are "foreshortened" and difficult to estimate even by a well experienced operator. Under certain conditions, the image seen by an operator in the eyepiece of the borescope may be reversed and/or inverted. Under such conditions, inspections of the ID surface of a round or symmetrical tube makes it difficult to establish a "frame of reference" for locating and identifying surface defects.

Despite the difficulties and shortcomings of visual inspection equipment, visual inspection is still the most important in the PRTR. At present, the use of the CCTV has been discontinued because of operator inexperience and excessive maintenance. Borescope examinations are still continuing at the rate of about 80% or about 60 pressure tubes per year.

The visual inspection system designed for the N-Reactor uses a special high optical resolution CCTV which can be

inserted into the pressure tube.^(19,20) This CCTV system can optically resolve two 0.003 in. diameter wires spaced as close as 0.010 in. The optical system is equipped with a moveable mirror. This mirror, when at 45 degrees to the axis of the CCTV camera, provides a normal and enlarged view of the pressure tube wall. Surface defects, when viewed normally and obliquely, are more easily identified by even relatively inexperienced personnel.

The above CCTV system still has appreciably less optical resolving power than do monocular borescopes. Hence, at N-Reactor, the CCTV system is used primarily for general scanning of the ID surfaces of the pressure tube. A video tape recorder is also available to provide a permanent record of the ID surface condition of the pressure tube. Detailed examinations of selected areas within the tube are done with a monocular borescope.

CRNL personnel use a monocular borescope exclusively for visual examinations of the ID surface of pressure tubes.⁽²¹⁾ Relatively few visual inspections are made in the AECL in-reactor monitoring program. The overall monitoring effort to date is largely limited to one tube in NRX, three in NRU and one in NPD. Previous visual examinations revealed only nominal surface damage from fretting, and surface scratching by fuel element supports. A device similar in principal to a phonograph needle is used to "scan" the entire surface of the ID of these pressure tubes and measure the depth and contour of surface defects, scratches, and the like. Successive "scans" of the tubes inspected can be compared and only in those instances, where significant "roughness" (e.g., 5 mil deep gouges, scratches, etc.) or significant changes are noted, is the tube then visually inspected with the borescope.

In spite of all the shortcomings of both CCTV systems and borescopes for visual examination, visual examinations of pressure tubes continues. For the PRTR and the N-Reactor, optical systems have become the primary inspection tool. Based on such experience, it appears that optical inspection systems might well be reviewed with the aim of achieving significant improvements in them. As yet, there has been no effort to develop a binocular borescope which would provide a good three dimensional field of view. At present, CCTV systems do not provide adequate optical resolution, color, or simplicity in operation and maintenance. Any further effort with CCTV systems is probably not warranted until significant breakthroughs in the television industry come about.

Surface Defect Measuring Instrumentation

A modified standard dial gage is used for measuring surface defects such as local tube wall erosion by fretting corrosion, gouges, and scratches in the PRTR.⁽¹⁸⁾ The dial gage is mounted on the borescope within the normal field of vision through the borescope. The dial gage is mounted on a moveable piston which is actuated by air. When actuated, the dial gage moves toward the tube wall until a curved "shoe" fitted to the stem of the dial gage makes contact with the tube wall. The contact point of the dial gage extends through the "shoe" until it contacts the "bottom" of the surface depression, scratch, etc. When repeated at a point on the tube wall adjacent to the defect, the dial indicator gives a different reading and the difference denotes the depth of the scratch, gouge, or other surface defect. This device has worked satisfactorily for measuring fretting corrosion erosion, gouges, and open "blister" defects on the ID surface. The dimensions of the contact point preclude depth measurements of scratches 10 mils or less in width.

The device used in the N-Reactor equipment^(18,19) is very similar to that used in the PRTR. In this case, actuation of the dial gage is through an electric motor driven cam rather than by air.

The device developed by CRNL is quite similar in principle to a phonograph needle.⁽²¹⁾ Actually three "needles" are used to cover the expected range of defect magnitudes. The gage is spiraled along the tube so as to provide a complete surface scan of the ID surface of the tube. Defect measurements are recorded on a strip chart. Reportedly, this gage has worked satisfactorily for the past several years.

Dimensional Gaging Instrumentation

The inside diameters of the pressure tubes in the PRTR were measured at two mutually perpendicular axes along the full length of the 3.25 in. bore by means of two LVDT systems.⁽⁷⁾ The LVDT gages are calibrated and checked before and after measuring each tube. Subsequent measurements were to be made at the same perpendicular axes for comparison purposes. Any diameter changes would provide the measure of creep deformation that had occurred between measurements intervals. In theory, this scheme appeared to be good. It was difficult, however, to obtain subsequent measurements in exactly the same planes of the original measurements. If the pressure tubes had been perfectly round, there would be no difficulties. But since all tubes are randomly out-of-round to some degree or other along their length, some measurement error was introduced. This error was estimated to be about 0.1%. After about 3 years of measurements no deformation significantly greater than the error measurement was observed. (Subsequent diameter measurements obtained with a Pi tape on tubes removed from the PRTR showed approximate diameter increases of between 0.1 and 0.2%.) In-reactor diameter measurements have been discontinued for the past several years. Future diameter measurements are expected to be obtained only from tubes removed from the reactor and by means of a Pi tape. Pi taping can be done in the storage basin with little effort and expense.

The ID gage for N-Reactor pressure tubes is identical with the LVDT system initially used in the PRTR.⁽¹⁷⁾ To date no measurements have been made.

The gage developed by CRNL is of the LVDT type similar to that used in the PRTR but with provision for measuring three equally spaced diameters.⁽²¹⁾ The LVDT gages are calibrated and checked before and after each tube is measured. These measurements are combined to provide an average diameter at specific distances along the length of the tube. These techniques have been useful in detecting flux influenced pressure tube creep in the test loops at NRX and NRU reactors. Measurements of the inside diameters of the pressure tubes in the NRX and NRU test loops are continuing.

Gas Gap Measuring Gage for the PRTR

The PRTR uses flowing helium within the annulus between the OD of the Zircaloy-2 pressure tubes and the ID of the aluminum calandria tube as a heat transfer barrier between the hot primary coolant system and the cold D₂O moderator system. Sufficient bowing of either the Zircaloy-2 or aluminum tubes to cause contact of the two tubes could cause boiling in the moderator. Using four eddy current coils spaced at 90 degree intervals, the eccentricity of the gas annulus could be measured.^(18, 22-24) The eddy current coils are held in contact with the ID surface of the Zircaloy-2 pressure tube. Using an appropriate exciting frequency, the coil can detect the distance it is away from the aluminum calandria tube. The device must be calibrated and checked before and after measuring each tube. Relatively little bowing was observed. The gage as built could only be used when the reactor was shutdown; hence, no information under actual operation was obtainable. This gage is no longer used but has been maintained on a standby basis.

Hydride Detection Instrumentation

To date, a really usable hydride detection instrument has not been developed. Both eddy currents and ultrasonic can be made to respond to the presence of large concentrations of zirconium hydride. However, both types of instruments respond to other changes in metallurgical structure (e.g., grain size, orientations, etc.). For this reason, it is quite difficult to interpret any signal obtained from either type of instrument.

An ultrastable eddy current device that could detect hydride concentrations as low as 250 ppm was demonstrated in the laboratory.⁽²⁵⁾ This device was used in the K reactors at Hanford, with rather indifferent success.^(26,27) A prototype ultrasonic device was built for use in N-Reactor, but was never used because of mechanical problems.⁽²⁸⁾

Tubes removed from the PRTR are examined metallographically for reasons other than hydride detection. As a result, adequate data were obtained without the use of an in-reactor gage.

CRNL has done little or no work toward the development of a hydride detection gage.

Over the past several years, laboratory investigations have included the effects of hydriding on the general mechanical properties. While entirely adequate knowledge concerning the effect of hydriding on pressure tube properties is not yet available, work is proceeding in that direction. Hence, the incentive for a hydride detection instrument has lessened to some extent.

Wall Thickness Measurements

Although wall thickness measuring prototypes were developed for PRTR pressure tubes and the pressure tubes in NRX and NRU, wall thickness measuring gages have not been used on pressure tubes in situ. The generally good performance of Zircaloy-2

pressure tubes and the conservatism existing in the design of the tubes now in the various reactors seems to eliminate any need for wall thickness measurements.

Ultrasonic methods requiring good coupling between the transducer and the tube wall were used in the prototype gages. For this reason, measurements are most easily obtained when the tube is full of water. This is in sharp contrast to all the other inspections which require that the tube be empty and dry. Potential D₂O losses also discouraged use of ultrasonic wall thickness measuring gages.

10.5 SURVEILLANCE AND TUBE REMOVAL CRITERIA GRID

A surveillance criteria and tube removal criteria grid have been prepared covering the operational standards and surveillance for the PRTR and N-Reactor.^(3-6,29) A column called "Comments and Priorities" is included that gives an evaluation of existing specifications and shows where additional specifications are needed. The specifications are rated as follows:

- I. Areas where ASTM specifications and other existing specifications and operating or maintenance standards are adequate.
- II. Areas where existing ASTM specifications and operating or maintenance standards should be modified.
- III. Areas where no specifications operating or maintenance standards exist and are needed.

Items, believed necessary to assure quality of pressure tubes are classified I, II or III as defined above. Also, priorities are listed for the R&D efforts needed to modify or establish, where absent, specifications, operating and/or maintenance standards for pressure tubes in heavy water reactors. These priorities are defined as follows:

- A. Areas which have a major effect on tube life, reliability or operating and maintenance procedures, which are not backed up by common commercial practice or other specifications.

- B. Areas which have a major effect on tube life, reliability or operating and maintenance procedures but which are backed up substantially by other specifications, operating or maintenance standards.
- C. Areas which have a minor effect on tube life, reliability or operating and maintenance procedures and for which adequate technology exists.

TABLE 10.1. Surveillance and Tube Removal Criteria

Surveillance Criteria

A. Operating Incidents

Item	PRTR	N-Reactor	Comments and Priorities
1) Rupture of a fuel element	BNWL-MA-1. Visual inspection in situ if fuel element inspection in storage basin shows evidence of contact with tube. If tube examination shows evidence of damage, tube is removed, inspected and proof tested before returning to service.	(Tentative) Visual inspection in situ if there is evidence that ruptured fuel element contacted tube surface. Contact is assumed if: 1) 5 times greater than normal force is required to discharge fuel element. 2) If rupture occurred in an outer fuel element.	II-A. An NDT test for hydriding would be desirable. More data and experience are desirable to establish actual service limiting damage.
2) Excessive temperature excursion of a pressure tube	BNWL-MA-1. If temperature of tube exceeds 600 °F, it must be removed, inspected and proof tested before returning to service if undamaged.	(Tentative) In situ inspection required if tube has been subjected to prolonged hot channel service. Temperature and time unspecified.	II-A. BNWL-MA-1 appears to be unnecessarily stringent. For N-Reactor there are no provisions for spotting a "hot" coolant channel by operating personnel.
3) Loss of flow in a pressure tube	Pressure drop limits are set to scram the reactor. Fuel elements examined to determine cause of flow loss.	Pressure drop limits are set to scram reactor. Fuel elements examined to determine cause of flow loss.	I. Principally a problem in operating procedure.
4) Continued high concentration levels of Zr corrosion product in primary coolant	No operating standards	No operating standards	II-B. Appreciable efforts are needed to establish a monitoring method and concentration levels of Zr corrosion product in the primary coolant which can reliably signal the need for fuel element and pressure tube inspections.
5) Prolonged vibrations of one or more pressure tubes	No operating standards	No operating standards	III-B. Some work has been done under PRP Program to correlate vibration frequency and amplitude with fretting. Work also needs to be done to ascertain the effects of superimposing pressure cycling and bending from flexural vibrations on fatigue crack growth rates.

10.24

BNWL-746

TABLE 10.1. Surveillance Criteria (contd)

B. Inspection Equipment

Item	PRTR	N-Reactor	Comments and Priorities
1) Visual	Borescope + CCTV and direct view - Monocular CVTR closed circuit TV	Borescope and/or CCTV Monocular	II-B. Development of binocular may be quite desirable. BNW has technology and personnel. Holography may also be applicable
2) Flaw detection (Surface and internal)	None used other than visual for surface flaws	None used other than visual for surface flaws	III-A. Need to develop instruments which are easily and reliably used by semi-skilled operators. Basic technology is available.
3) Hydride detection (materials evaluation efforts have not progressed to point where "go-no-go" limit can be set.)	None used now	None used - an eddy current device is available	III-A. Reliable and simple-to-use equipment is not yet available. Development work has stopped. Neutron radiography reported as a potential detection method.
4) Dimensional measurements	Routine measurements discontinued. Pi taping done on a convenience basis on tubes removed from the reactor.	ID measurements obtained for 2 to 5% of pressure tubes in the reactor	I. Technology is available. Design of equipment probably needs to be tailored to each reactor.

10.25

BNWL-746

TABLE 10.1. Surveillance Criteria (contd)

C. Inspection Schedules and Frequency

Item	PRTR	N-Reactor	Comments and Priorities
1) Visual	80% of the 85 pressure tubes inspected once per year. Special tests require inspection at specified intervals. "Operating incidents" require inspection.	Minimum of 2% and up to 5% of the 1,000 pressure tubes in the core. Operating incidents require inspections.	I. For PRTR inspection frequency probably exceeds needs. For N-Reactor, there is not enough experience to assess adequacy of current schedules.
2) Flaw detection	Surface flaw detection is incorporated with visual inspection. NDT methods not used.	Surface flaw detection is incorporated with visual inspections. NDT methods not used.	II-A. Visual methods are not adequate to detect fine type cracks.
3) Hydride detection	Not scheduled. Operation depends on examination and destructive testing of tubes permanently removed from reactor.	Not scheduled. Same as for PRTR. An NDT instrument is available for use.	II-A. Periodic tube removal and destructive examination is unsuitable for commercial reactors in most cases.
4) Zr corrosion product. Content of primary coolant	Sampling discontinued. Cost too great and technique is inadequate to detect corrosion products	Not scheduled	III-B. Feasibility of on line monitoring should be undertaken.

10.26

BNWL-746

TABLE 10.1. Tube Removal Criteria

Item	PRTR	N-Reactor	Comments and Priorities
1) Overtemperature	Tube removed for out-of-reactor inspection if tube temperature exceeds 600 °F.	Action is based on a review by Plant Engineering and by Research and Engineering Groups	II-C. PRTR standards appear to be overly conservative. Limit for inspection may be more realistic than one for mandatory removal.
2) Dimensions (Creep, straightness)	No operating standards which specify limit of dimensional changes for mandatory removal.	Tentative deformation limit is a 2% increase in the tube ID	II-A. In-reactor creep and stress rupture data is needed to establish a realistic deformation limit and to establish appropriate working stress levels and minimize in-reactor inspections. Straightness requirements should be determined by designer.
3) Flaw Size (Surface and internal which may enhance probability of "brittle" failure)	No operating standard for internal flaws. Tentative standards based on a safety factor of 4 are: 1) failure - flaw length, 1 in. - flaw depth, 1/8 in. 2) leakage - flaw length, 1/4 in. - flaw depth, 1/8 in.	No standard	II-A. For PRTR, the tentative flaw size specification is based on unirradiated tests. Irradiated data is required to assess the effects of: 1) neutron exposure 2) hydriding 3) combined reactor service environment on the fracture toughness of pressure tubes.

TABLE 10.1. Tube Removal Criteria (contd)

<u>Item</u>	<u>PRTR</u>	<u>N-Reactor</u>	<u>Comments and Priorities</u>
4) Neutron exposure (cumulative)	No standard	No standard	III. There is no information which as yet can suggest whether or not a neutron exposure limit exists.
5) Hydrogen uptake from corrosion or other sources	No standard	Tentative limit of 1,000 ppm locally. Unspecified as to how measured.	II. As yet there is no reliably usable instrument for measuring, hence limit is somewhat unrealistic. Effects of hydriding on fracture toughness and other pertinent mechanical properties needs further investigation before a reasonable limit is established. Hydrogen uptake from corrosion can be estimated. From a fuel failure or other source, the extent of hydriding cannot easily or reliably be estimated.

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11.0 RELIABILITY AND SENSITIVITY ANALYSIS

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11.1 RELIABILITY ANALYSIS

The reliability of a device or system is the probability that it will perform in a specified manner for a given period of time when used in a particular environment. The purpose of a reliability program for the HWR pressure tubes is to assure a high probability that the zirconium alloy pressure tube assembly will operate without failure over the service life of the reactor (e.g., 30 years).

Reliability studies on reactor pressure tubes are almost nonexistent in the literature. Most of the reliability studies that have been done on nuclear systems concern reactor safeguards rather than specific components. However, four reports relating directly to pressure tube reliability have been located. One of the reports obtained was a study to determine test program requirements for demonstration of pressure tube assembly reliability for the Heavy Water Organic Cooled Reactor (HWOCR) program.⁽¹⁾ The second report is the result of a study to formulate recommendations for conducting analytical and experimental work to support a firm assessment of reactor plant piping reliability, and to allow upgrading of design codes where necessary.⁽²⁾ The third and fourth reports are concerned with the development of reliability models and procedures which can be used to estimate the reliability of primary systems of specific reactors. Efforts to identify the failure-causing factors and the development of the probability equations are discussed.^(3,4)

To be useful for reliability analysis, data on pressure tube failure, life tests, and operating experience must document failure modes, causes of failure, stresses at failure, time to failure, and environmental effects. The available data in the literature do not meet these criteria.

11.2 SENSITIVITY ANALYSIS

A sensitivity analysis is a method of ranking the various input parameters to a design problem according to their influence on specified criteria, and according to the subsequent errors produced in the criteria due to uncertainties on the parameters. This analytical procedure is used to recommend the best way resources should be expended to reach a set of objectives. A sensitivity analysis of the design of pressure tubes for the heavy water reactor zirconium alloy pressure tube program would reveal those critical areas in the program where the greatest amount of effort and resources should be concentrated to assure that the pressure tubes will operate without failure over the planned service life of the reactor.

Only two references to sensitivity analysis on pressure tube design were found in the literature. One report⁽³⁾ considers the parameter influences in the synthesis of a sample reliability model for a reactor piping system. This report deals only with showing how reliability engineering can be used in estimating the probabilities of failures and does not actually analyze the pressure tubes. The second report⁽⁴⁾ discusses system reliability analysis and introduces the concept of "sensitive factors."

A sensitivity analysis is useful in insuring that the most influential parameters are considered. This is done by an iterative process in which the mathematical models and all facets of the overall sensitivity analysis are improved throughout the course of the investigation. Parameters which may be influential in an early part of the analysis may be replaced by other parameters as successively better information is generated. Better values of the parameters, better information that will alter the system model, etc. are factored into successive analyses to continually upgrade the quality of the answers.

There is relatively little information available on reliability and sensitivity analysis for the design of reactor process tubes. There is some difficulty in obtaining information for these analyses because there have not been sufficient failures from which to gather information, thus information from laboratory testing has to be substituted.

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12.0 OTHER PRESSURE TUBE MATERIALS

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12.1 PROPERTIES OBTAINED FROM COUPONS

Tensile Strength

Zircaloy-4

Bement⁽¹⁾ has summarized the effects of hydriding on zirconium. In brief, oriented hydrides provide planes of weakness which embrittle zirconium alloys. The removal of nickel from Zircaloy-2 and increasing the iron content to compensate for removal of nickel produces the alloy Zircaloy-4. The rate of hydrogen pickup is significantly reduced in Zircaloy-4. As might be expected, the alloy response to irradiation shown in Figure 12.1 is not significantly different from Zircaloy-2. The variation in tensile properties of cold rolled and stress relieved Zircaloy-4 plate with test temperature and irradiation is given in Table 12.1.⁽²⁾

Zr-3 Nb-1 Sn

The Zr-3 Nb-1 Sn alloy has a higher strength than Zircaloy-2. Both alloys are nearly equivalent with respect to corrosion behavior in steam. Irradiation of the Zr-Nb-Sn alloy produces, initially, a drop of several percent in elongation at both room temperature and 300 °C.⁽³⁾ The effect, however, seems to saturate above a fluence of 1×10^{19} n/cm² ($E > 1$ MeV) with minimum ductility of 10%. Irradiation also causes an increase in strength values of approximately 50%. On the average, the Zr-3 Nb-1 Sn alloy, after irradiation, shows a few percent higher strength and a few percent lower ductility than Zircaloy-2.

FIGURE 12.1. Comparison of Effects of 100 °C Irradiation on the Room Temperature Tensile Properties of Zircaloy-2 and Zircaloy-4

TABLE 12.1. The Effect of Irradiation on the Tensile Properties of Zircaloy-4 Plate Material Cold Rolled 70% and Stress Relieved at 950 °F for 4 hr. (All Tests Performed at a Strain Rate of 1.5 in./in./hr)

Specimen No.	ETR Loop	Loop Temp., °F	Total Exposure, nvt (E > 1 MeV)	Test Temp., °F	0.2% Yield Strength, psi	Ultimate Tensile Strength, psi	Elongation in 2 in. ^(a)		Change in Yield Strength, psi	Expected Change in Yield Strength, psi (Eq. 35)
							Uniform, %	Total, %		
82-85	--	--	0	RT	87,800	93,400	3.0	13.0		
82-84	L-12	500	1.7×10^{20}	RT	113,900	114,600	0.5	1.5	26,100	25,000
82-91	--	--	0	560	59,300	64,400	2.0	11.0		
82-83	L-12	500	1.8×10^{20}							
	L-8	430	5.0×10^{20}	560	69,400	71,200	1.0	5.0	10,100	38,500
82-86	--	--	0	700	53,200	56,700	1.3	9.5		
82-90	L-12	500	1.7×10^{20}	700	65,300	66,700	1.0	5.0	12,100	25,000

(a) From the load-extension autographic recording.

Zr-2.5 Nb

Zirconium-niobium alloys are stronger than Zircaloy-2 even when given heat treatments to optimize corrosion properties at the expense of mechanical strength. Beta-quenched and aged Zr-2.5 Nb is about 50% stronger than Zircaloy-2.⁽¹⁾

Ells and Fidleris^(4,5) at Chalk River described the relation between the metallurgical condition and response of tensile properties of the alloy to neutron irradiation. Their results for the ($\alpha + \beta$)-quenched and aged material compared to the β -quenched and aged material are shown in Figures 12.2 and 12.3. The response of the alloy was also compared in the slow cooled rather than quenched condition (Figures 12.4 and 12.5). The authors concluded that the Zr-2.5 Nb alloy quenched from the ($\alpha + \beta$)-phase and aged 24 hr at 500 °C undergoes very small losses in fracture ductility from neutron irradiation and is thus one of the optimum conditions for the alloy.

G. W. Parry, also of Chalk River, has investigated the effect of irradiation on the tensile properties of Zr-2.5 Nb tubing.⁽⁶⁾ The usual increases in strength and decreases in ductility to about 50% of the unirradiated values were observed. A greater percent increase in yield strength is seen in longitudinal specimens than in transverse specimens.

One set of fatigue results has been obtained on zirconium or its alloys.⁽⁷⁾ Crystal bar zirconium and a Zr-2.5 Sn alloy were irradiated at 280 °C (540 °F) to 1 to 6×10^{19} nvt. Unnotched and notched specimens taken in the longitudinal direction from rolled plate were tested in bending at room temperature. Irradiation had no significant effect on the S-N curve, in either notched or unnotched conditions.

FIGURE 12.2. Engineering Stress-Strain Curves for Zr-2.5 wt% Nb Alloy Quenched from ($\alpha + \beta$)-Phase and Aged 24 hr at 500 °C; T Material; Irradiation at 250 to 325 °C to 5.4×10^{20} n/cm². Tests at 300 °C

FIGURE 12.3. Engineering Stress-Strain Curves for Zr-2.5 wt% Nb Alloy Quenched from β -Phase and Aged 24 hr at 500 °C; AW Material; Irradiation at 250 to 325 °C to 3.0×10^{20} n/cm². Test Temperatures Shown on Curves.

FIGURE 12.4. Engineering Stress-Strain Curves for Zr-2.5 wt% Nb Alloy Slow Cooled from ($\alpha + \beta$)-Phase; AW Material; Irradiation at 250 to 350 °C and Tested at 300 °C. Integrated Flux Shown on Curves.

FIGURE 12.5. Engineering Stress-Strain Curves for Zr-2.5 wt% Nb Alloy Slow Cooled from the β -Phase; AW Material; Irradiation at 250 to 325 °C. Tests at 300 °C

CreepZircaloy-4

Postirradiation creep tests have been performed on Zircaloy-4 which have been irradiated to fast fluences up to 2.5×10^{21} nvt.⁽⁸⁾ Tests were performed at 285 °C with a stress of 20,000 psi. The tests were only conducted to 100 hr. Since the creep rate for Zircaloy-4 at this stress and temperature is very low, only 0.1% creep or less was observed. With this very small amount of creep, it is difficult to make any conclusions from these results other than the irradiation did not cause a large increase in creep rate. These results are shown in Table 12.2.

TABLE 12.2. Summary of Pre- and Post-Irradiation Creep of Zircaloy-4 at 550 °F and 20,000 psi

Condition	nvt (E > 1 MeV)	% Creep Strain		
		1 hr	10 hr	100 hr
Annealed	None	0.10	0.11	0.11
	1×10^{21}	0.04	0.05	0.05
	1.4×10^{21}	0.07	0.08	0.08
	2.5×10^{21}	0.06	0.07	0.07
	2.5×10^{21}	0.10	0.10	0.11
	None	0.08	0.09	0.10
As-Extruded	1.0×10^{21}	0.07	0.08	0.08
	1.0×10^{21}	0.06	0.06	0.06
	1.4×10^{21}	0.03	0.04	0.04
	1.4×10^{21}	0.05	0.06	0.06
	None	0.06	0.07	0.07
	2.5×10^{21}	0.05	0.05	0.05
As-Extruded (100 ppm H ₂)	2.5×10^{21}	0.08	0.08	0.09

Zr-2.5 Nb

In-reactor creep of Zr-2.5 wt.% Nb is being studied at Pacific Northwest Laboratory,⁽⁹⁾ but the results have not yet been published. No significant differences are being found in creep rate for temperatures of 350, 375, and 400 °C. The

in-reactor creep rates are significantly higher at 300 °C and stresses of 36,500, 45,000, and 55,000 psi and at 320 °C and 55,000 psi than the corresponding unirradiated control creep rate. In-reactor creep studies are being conducted at AECL⁽¹⁰⁾ on Zr-2.5 wt.% Nb which show faster in-reactor creep rates during irradiation than the out-of-reactor creep rates.

In-reactor creep tests on pressurized tubes of Zr-2.5 wt.% Nb and Zircaloy-4 have been reported⁽¹¹⁾ to have creep rates accelerated by neutron irradiation. The Zr-2.5 wt.% Nb was tested at stresses of 15,500 and 21,000 psi at 285 °C. The Zircaloy-4 was tested at 5000 psi and 300 to 345 °C.

Fe-Base Alloys

The Fe-Cr-Al alloys are of interest for water reactors because of their good corrosion resistance, low capture cross section relative to the nickel alloys and the 300 series stainless steels, and their superiority in mechanical properties over the regular 400 series stainless steels.

The combined effects of irradiation and annealing on several Fe-Cr-Al base experimental alloys are depicted in Figure 12.6.^(12,13) The effects of 650 °C irradiation upon the room temperature properties of selected iron-base alloys are reported by Claudson and Pessl and shown in Table 12.3.

An interesting variation of the Fe-Cr-Al system is an alloy containing yttrium. Additions of less than 1% yttrium has markedly improved the oxidation resistance of this family of alloys.⁽¹⁴⁾ In addition, two alloys have been reported by Wukusick⁽¹⁵⁾ of General Electric as optimum; Fe-25 Cr-4 Al-1 Y (the 2541 alloy) for high temperature oxidation resistance, and Fe-5 Cr-6 Al-1 Y for low temperature stability. The alloys can also be prepared by an atomized powder process realizing improved strength and ductility through a finely dispersed

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TABLE 12.3. Effects of Neutron Irradiation in Contaminated Helium Atmosphere at 650 °C (1202 °F) Max. upon the Room Temperature Hardness and Tensile Properties at Atmospheric and Elevated Temperature of Some Iron-Base Alloys

Alloy	Unirradiated				Irradiated			
	Hardness, ^(a) R _B	YS 1000 psi	US psi	Elong., %	2.53 x 10 ¹⁹ nvt (E > 1 MeV)			
					Hardness, ^(a) R _B	YS 1000 psi	US psi	Elong., %
Tested at Room Temperature								
AISI 316 SS	66	30	81	62	63	27	78	56
AISI 347 SS	76	53	100	44	72	40	95	37
AISI 406 SS	87	67	89	23	--	70	91	25
Ferral Mod.	95	74	98	16	95	77	77	3
Tested at 700 °C (1292 °F)								
AISI 316 SS	67	13	36	48	66	25	67	46
AISI 347 SS	75	33	43	34	74	39	68	27
AISI 406 SS	86	14	14	74	76	31	42	54
Ferral Mod.	90	16	19	53	100	42	54	29

(a) Hardness tested at room temperature.

Y Fe₉ phase. General Electric has reported a limited amount of irradiation effects data on a Fe-15 Cr-4 Al-1 Y alloy. These data are shown in Table 12.4.⁽¹⁶⁾

TABLE 12.4. Strain-Rate Sensitivity of Irradiated^(a) and Unirradiated^(b) Fe-15 Cr-4 Al-1 Y Alloy

Deformation Temperature, °C	Strain Rate, %/min	Strength, 1000 psi		Ductility, %	
		0.2% Offset Yield	Engineering Ultimate	True Uniform Strain	Total Elongation
500	20.0	32.2(28.0)	45.4(37.9)	8.8(8.1)	22.5(17.9)
	2.0	34.6(30.7)	41.6(34.5)	10.8(4.2)	14.9(19.7)
	0.2	30.8(20.1)	35.8(24.8)	6.0(7.1)	23.2(26.3)
600	20.0	29.7(24.4)	34.8(28.1)	6.3(4.1)	32.7(25.4)
	2.0	27.2(19.3)	29.9(21.4)	4.5(4.3)	30.3(31.1)
	0.2	21.8(15.3)	22.8(17.1)	4.0(3.5)	30.4(27.2)
700	20.0	18.5(13.8)	20.3(14.6)	6.5(2.6)	39.0(62.0)
	2.0	14.2(9.8)	15.6(9.9)	4.4(3.2)	51.9(51.0)
	0.2	11.4(8.2)	12.3(8.5)	2.6(2.0)	37.8(44.3)
800	20.0	12.0(7.6)	12.4(7.9)	4.4(2.3)	50.9(75.7)
	2.0	7.9(4.3)	8.1(4.5)	3.5(3.3)	64.0(74.1)
	0.2	3.9(3.5)	4.7(3.7)	1.9(1.8)	78.1(76.4)
871	20.0	6.2(6.8)	6.5(6.9)	2.1(1.0)	58.3(82.5)
	2.0	4.3(4.0)	4.8(4.2)	2.6(3.3)	76.4(79.3)
	0.2	2.8(2.2)	3.2(2.5)	3.5(4.1)	93.1(92.7)

(a) First entries are data from samples irradiated at 50 °C to a thermal fluence of 1×10^{20} n/cm² and a fast fluence of 1.5×10^{19} n/cm² ($E > 1$ MeV).

(b) Values given in parentheses.

Of possible interest is the Type 410 SS alloy. This alloy, ferritic in the annealed condition, can be hardened to a martensitic structure. Studies of irradiation damage to this material has been reported by Lovell.⁽¹⁷⁾

The strength and the ductility for room temperature tests versus fast neutron fluence for 60 and 290 °C irradiations of annealed Type 410 SS are shown in Figures 12.7 through 12.10. The strength data show the 290 °C irradiations to be continuing to increase in strength at 6×10^{21} , whereas the 60 °C irradiations show no increase in strength from 1×10^{20} to the

12.12

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FIGURE 12.7. The Effect of Neutron Exposure at 60 °C on the Room Temperature Strength of Annealed AISI 410 SS

FIGURE 12.8. The Effect of Neutron Exposure at 60 °C on the Room Temperature Ductility of Annealed AISI 410 SS

FIGURE 12.9. The Effect of Neutron Exposure at 290 °C on the Room Temperature Tensile Strength of Annealed AISI 410 SS

FIGURE 12.10. The Effect of Neutron Exposure at 290 °C on the Room Temperature Ductility of Annealed AISI 410 SS

highest fluence (5×10^{21}). The ductility at both irradiation temperatures are very low above a fluence of 2×10^{20} .

The effect of test temperature on specimens irradiated at 60 °C can be illustrated by the data from the highest fluence (5.21×10^{21}). The strength value (Figure 12.11) for specimens tested at 450 and 600 °C are the same as for unirradiated specimens. The ductility as shown in Figure 12.12 has also recovered in tests at 450 and 600 °C.

The marked difference between irradiation at 60 and 290 °C is illustrated by the composite plot of strength versus test temperature for various irradiations at 290 °C (see Figure 12.13). The irradiation hardening is not removed from specimens irradiated at 290 °C then tested at 450 and 500 °C. The hardening is present until the 600 °C test temperature, where the recovery of unirradiated properties is complete. The 290 °C irradiation then produces irradiation defects that are more effective in hardening the annealed Type 410 SS than irradiations at 60 °C. The defects are also more stable in that they are effective in hardening at higher test temperatures for a given fast neutron fluence. Of particular interest is the total recovery of ductility at 600 °C test temperature (see Figure 12.14).

12.2 PROPERTIES OBTAINED FROM PRESSURE TUBING

Scope of Effort

The ultimate burst strength, creep and stress rupture strength, fatigue crack growth, and crack propagation properties of Zr-2.5 wt% Nb alloy tubing have been under investigation for the past several years. There is appreciable interest in this alloy, particularly by AECL, for pressure tubes in water cooled power reactors now in design and/or construction stages.

FIGURE 12.11. The Effect of Test Temperature on the Ductility of Annealed 410 SS Irradiated at 60 °C to $5.21 \times 10^{21} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$)

FIGURE 12.12. The Effect of Test Temperature on the Strength of Annealed 410 SS Irradiated at 60 °C to $5.21 \times 10^{21} \text{ n/cm}^2$ ($E > 1 \text{ MeV}$)

FIGURE 12.13. The Effect of Test Temperature on the Strength of Annealed AISI 410 SS Irradiated at 290°C

FIGURE 12.14. The Effect of Test Temperature on the Ductility of Annealed AISI 410 SS Irradiated at 290 °C

While there are significant amounts of data from unirradiated material, very little data have been obtained from irradiated tubing. R&D efforts are underway to acquire such data. These efforts, however, are appreciably less than are those for Zircaloy-2 and the progress of these R&D efforts have been delayed because of the unavailability of irradiated tubing. Several test pieces are currently being irradiated in the PRTR; prototype pressure tubes have also been installed in test loops in the NRX and NRU; one tube is also operating as a standard process channel in the PRTR [see Table 12.5⁽¹⁸⁾].

Burst and crack propagation tests have been done on prototypic Zr-2.5 wt% Nb alloy tubing in the 30 and 60% cold-worked conditions and the heat treated conditions. Tubing was nominally 2.1 in. ID by 0.170 in. wall thickness. Except for tests on hydrided material, the tubular test specimens were not autoclaved. The heat treatments consisted of:

- 880 °C solution treatment and water quench.
- 880 °C solution treatment, water quench and age 24 hr at 500 °C.
- 880 °C solution treatment, water quench and 15% cold work.
- 880 °C solution treatment, water quench, 15% cold work and age 24 hr at 500 °C.

Of all the above heat treatment, the last one probably represents the best overall heat treatment.

Ultimate Strength of Tubing

Ultimate strength of tubing is determined by burst tests. In burst tests, a length of tubing (length is about 10x diameter) is internally pressurized without axial restraints until failure. Under these conditions, the ratio of hoop to axial stresses is 2 to 1.

TABLE 12.5. *In-Reactor Tests of Zr-Nb Alloy Pressure Tubes*

Tube	Material	Stress, psi	Temp., °C	Operating Time, hrs	Exposure 10 ²⁰ n/cm ² (>1 MeV)	Remarks
U1MkV-1	CW Zr-2.5 Nb	15,500 21,000	285 285	6,850	8.5	Final results in Exp-NRU-10406
U1MkV11	CW Zr-2.5 Nb	16,500 22,000	270 270	4,040	4.6	
PRTR	CW Zr-2.5 Nb	19,000	250-270	~500	~1	
X5Mk1V	h.t. Zr-2.5 Nb	26,000	240-250	~500	~0.2	Installed - May 1967
X5Mk11	h.t. Zr-2.5 Nb	19,000	240-250	14,300	3	Removed - May 1967
U2Mk1X	CW Zr-2	13,400 17,300	270-292	3,350		Removed - June 1967
NPD	CW Zr-2	10,500	252-273	29,000	12	
PRTR6090	CW Zr-2	11,000	250-270	11,000	21	Reference gauged Sept 1966
PRTR6065	CW Zr-2	11,000	250-270	11,000	26	Reference gauged Sept 1966
U3MkV-1	CW Zr-4	6,500 to 4,300	300-345	13,000	15	

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For comparison purposes, uniaxial tensile properties obtained from coupons cut from pressure tubing is included and shown in Tables 12.6 and 12.7.⁽¹⁹⁾ These data show the tensile strength of Zr-2.5 wt% Nb alloy to be significantly greater than that of Zircaloy-2. Tables 12.8 through 12.11 show the ultimate hoop strength of Zircaloy-2 and Zr-2.5 wt% Nb as determined from burst tests. Data in Tables 12.8, 12.10, and 12.11 were obtained from tubing with diameters of about 2.38 to 3.2 in. and wall thickness of 0.15 to 0.26 in. and R to t ratios of 6 to 7. Cold work tempers range from 18 to 50% CW. Data in Table 12.9 were obtained from 5.14 in. ID tubing with wall thickness of 0.12 and 0.2 in. and R to t ratios of 14 and 22. For Zircaloy-2, the UBS at room temperature ranges from about 90 to 95 ksi for annealed⁽²⁰⁻²²⁾ to about 115 ksi for cold-worked tubing.⁽²³⁾ The Zr-2.5 wt% Nb tubing ranges from a low of about 100 to about 125 ksi for cold worked and for heat treated. As temperature increases to 300 °C, Tables 12.8 and 12.9 show the difference in strength increases.

At room temperature, comparisons of uniaxial tensile with ultimate burst strengths show that cold-worked Zircaloy-2 tubing tends to benefit more from anisotropic effects than does Zr-2.5 wt% Nb alloy tubing. At 300 °C, the ratio of burst strength to ultimate tensile strength is still about the same. The effect of anisotropy, however, does not overcome the strength advantage offered through heat treatment of the Zr-2.5 wt% Nb alloy.

Fracture and Fatigue Properties

Appreciable efforts have been directed toward investigation of crack propagation characteristics including fatigue crack initiation and growth.^(19,24) In many of the crack propagation tests, defects were simulated by a milled slot cut

TABLE 12.6. Typical Properties of Zircaloy-2

Material Condition	Test Direction	Room-Temperature Properties				300 °C Properties			
		UTS, lb/in. ² x 10 ⁻³	0.2% PS, lb/in. ² x 10 ⁻³	Elong., % on 4√A	Reduction of Area, %	UTS, lb/in. ² x 10 ⁻³	0.2% PS, lb/in. ² x 10 ⁻³	Elong., % on 4√A	Reduction of Area, %
Annealed (Plate)	L	70	42	28	44	33	20	30	60
	T	72	60	25	52	35	24	26	70
20% Cold-Worked Tube (as drawn)	L	90	75	20	40	52	47	20	45
	T	87	78	20	40	50	46	20	45
20% Cold-Worked Tube (autoclaved)	L	84	63	20	45	49	45	20	45
	T	84	71	20	45	45	43	22	55
30% Cold-Worked Tube (as drawn)	L	101	72	20	35	58	56	20	54
	T	95	77	20	40	53	52	20	55
30% Cold-Worked Tube (autoclaved)	L	92	81	20	45	54	51	20	45
	T	89	81	20	42	50	48	20	55

TABLE 12.7. Typical Properties of Zr-2.5 Nb Alloy

Material Condition	Test Direction	Room-Temperature Properties				300 °C Properties			
		UTS, lb/in. ² x 10 ⁻³	0.2% PS, lb/in. ² x 10 ⁻³	Elong., % on 4√A	Reduction of Area, %	UTS, lb/in. ² x 10 ⁻³	0.2% PS, lb/in. ² x 10 ⁻³	Elong., % on 4√A	Reduction of Area, %
Annealed Zr-2.5 Nb (Plate)	L	85	81	25	65	45	28	30	65
	T	100	92	25	65	54	42	20	80
20% Cold-Worked Tube (as drawn)	L	105	80	18	45	70	-	20	40-50
	T	115	-	18	45	73	-	20	40-50
20% Cold-Worked Tube (autoclaved)	L	110	85	18	45	71	-	20	40-50
	T	120	95	18	40-50	75	-	20	60-70
Heat-Treated Tube (Q.880, T.500)	L	124	115	10	45	92	-	12	58
	T	128	120	10	35	93	-	10	43

Note: Autoclaving for 3 days at 400 °C has no significant effect on the tensile properties of annealed and heat-treated Zr-2.5 Nb alloy.

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TABLE 12.8. Burst Tests of Zircaloy-2 Pressure Tubing

Test	Temperature, °C	Ultimate Hoop Stress, psi	Total Elongation, %	Test	Temperature, °C	Ultimate Hoop Stress, psi	Total Elongation, %
N18B-1	302	61,300	22.0	K30B- 1	300	65,500	10.6
2	303	61,300	19.3	2	300	68,200	12.1
3	304	61,300	21.0	3	300	67,200	14.5
4	155	80,200	19.0	4	350	61,800	6.7
5	145	77,800	31.0	5	345	62,000	12.8
6	154	76,800	34.4	6	345	60,600	4.9
7	RT	105,000	21.0	7	154	86,000	18.3
8	RT	107,000	16.3	8	150	83,200	19.9
9	RT	107,000	13.2	9	157	84,000	25.1
N30B-1	RT	107,000	21.9	10	RT	115,000	8.4
2	RT	110,000	22.7	11	RT	117,000	12.6
3	RT	109,000	19.6	12	RT	116,500	11.5
4	150	80,500	34.2	K50B- 1	300	58,500	19.2
5	152	83,500	34.2	2	283	58,500	
6	150	82,300	34.2	3	270	59,200	
7	307	64,700	37.7	4	272	58,000	
8	304	61,500	35.5	5	150	80,700	19.2
9	305	64,300	35.8	6	150	81,700	15.8
N35B-1	295	66,400	20.8	7	RT	104,700	6.7
2	300	65,900	11.0	8	RT	107,200	12.5
3	300	64,300	21.3	9	RT	109,000	15.9
4	145	85,100	23.5				
5	150	82,800	20.0				
6	156	83,200	25.9				
7	RT	118,000	6.4				
8	RT	114,000	8.5				
9	RT	114,000	2.5				

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TABLE 12.9. 300 °C Tube Burst Properties

Material Condition	Hoop Stress at Failure, lb/in. ² x 10 ⁻³	Circumferential Strain at Burst, %	Maximum Reduction of Wall Thickness, %	Ratio Burst Stress/UTS (T)
20% Cold-Worked Zircaloy-2 Tube (autoclaved)	59	18-25	15-20	1.3
30% Cold-Worked Zircaloy-2 Tube (autoclaved)	67	~20	~20	1.34
20% Cold-Worked Zr-2.5 Nb Alloy Tube (autoclaved)	82	1-1.5	16-18	1.09
Heat-Treated Zr-2.5 Nb Tube (Q.880, T.550)	97	1.3-1.5	14-16	1.04

into the OD surface of the tube in a manner identical to that used with Zircaloy-2 work.^(21,25) Under these conditions, direct comparisons could be made. Tables 12.10 and 12.11 present the milled slot data for Zr-2.5 wt% Nb tubing in both the cold-worked and the heat-treated conditions. Comparisons between Zircaloy-2 and Zr-2.5 wt% Nb alloy tubing are shown in Figure 12.15. These data show that there is little if any real difference between the strength of defected tubing of both alloys. Neutron irradiation had little effect on Zircaloy-2. As yet there are no data for irradiated Zr-2.5 wt% Nb tubing. However, based on comparisons of irradiated versus nonirradiated tensile data for Zircaloy-2 and Zr-2.5 wt% Nb alloy,⁽²⁶⁾ it is anticipated that neutron irradiation will have little if any more effect than was observed in the case of Zircaloy-2 tubing with milled slots. (See Section 6, "Radiation Effects on Pressure Tube Properties.")

Some fatigue data have been obtained for Zr-2.5 wt% Nb alloy tubing.⁽²⁴⁾ Tubes were stressed by internal pressure in a tension-tension mode with the ratio of hoop to axial stresses of 2 to 1. Semicircular EDM (electrical discharge machined)

TABLE 12.10. Crack Propagation Tests of Cold-Worked Zr-2.5 wt% Nb

Test Temperature	Material Condition		Slot Length, in.	Hoop Strength, ksi	K _c (Calculated), ksi $\sqrt{\text{in.}}$	Failure Mode
	Cold Work	Hydrides				
Room Temp.	30%	None	None	114	---	Ductile
Room Temp.	30%	None	1/2	94	294	Ductile
Room Temp.	30%	None	1 1/2	42	238	Ductile
Room Temp.	30%	None	2 1/2	25	293	Ductile
Room Temp.	30%	None	4	26	619	Tension
Room Temp.	60%	None	None	121	---	Ductile
Room Temp.	60%	None	1/2	92	236	Ductile
Room Temp.	60%	None	1 1/2	41	229	Ductile
Room Temp.	60%	None	2 1/2	21	244	Ductile
Room Temp.	60%	None	4	22	516	Ductile
0 °C	30%	None	None	115	---	Tension
0 °C	30%	None	1/2	95	294	Ductile
0 °C	30%	None	1 1/2	43	251	Ductile
0 °C	30%	None	2 1/2	26.6	300	Ductile
0 °C	30%	None	4	28	669	Ductile
0 °C	60%	None	1 1/2	43	---	Ductile
-30 °C	30%	None	None	118	---	Tension
-30 °C	30%	None	1/2	92	245	Ductile
-30 °C	30%	None	1 1/2	43.5	246	Ductile
-30 °C	30%	None	2 1/2	29	343	Ductile
-30 °C	30%	None	4	32.5	785	Ductile
-30 °C	60%	None	None	127	---	Tension
-30 °C	60%	None	1/2	95	241	Tension
-30 °C	60%	None	1 1/2	40.5	220	Tension
-30 °C	60%	None	2 1/2	24	270	Tension
-30 °C	60%	None	4	14.2	329	Tension
150 °C	30%	None	None	85	---	Ductile
Room Temp.	60%	None	None	99.5	---	Ductile

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TABLE 12.10. (contd)

Test Temperature	Material Condition		Slot Length, in.	Hoop Strength, ksi	K _c (Calculated), ksi $\sqrt{\text{in.}}$	Failure Mode
	Cold Work	Hydrides				
300 °C	30%	None	None	84	---	Ductile
300 °C	30%	None	2 1/2	15.6	179	No Propagation
300 °C	60%	None	1/2	64	---	No Propagation
300 °C	60%	None	2 1/2	16	---	No Propagation
300 °C	30%	275 ppm H ₂	1/2	73	---	Tension
300 °C	30%	275 ppm H ₂	1 1/2	30	---	Tension
300 °C	30%	275 ppm H ₂	2 1/2	27	---	Tension
300 °C	60%	275 ppm H ₂	1/2	85	---	Tension
300 °C	60%	275 ppm H ₂	1 1/2	8.8	---	Tension
300 °C	60%	275 ppm H ₂	2 1/2	7.9	---	Tension
~50 °C	30%	275 ppm H ₂	1/2	26	---	Tension
150 °C	30%	275 ppm	1 1/2	26.3	---	Tension
150 °C	60%	275 ppm	1/2	34	---	Tension
300 °C	30%	275 ppm	1 1/2	34	---	No Propagation
300 °C	30%	275 ppm	2 1/2	8.2	---	No Propagation
300 °C	60%	275 ppm	None	87	---	Tension
300 °C	60%	275 ppm	1/2	43.5	84	No Propagation

$$K^2 \text{ (Calculated)} = \frac{\pi \sigma_f^2 C}{\cos \theta} \left(1 + \frac{5 \pi \lambda^2}{32} \right) \left(\frac{4 - \kappa}{2} \right)$$

where

$$\kappa = (3 - 4 \mu) / (1 + \mu) \text{ (Plane Stress Conditions),}$$

 μ = Poisson's Ratio,

C = 1/2 Slot Length,

R = Tube Radius,

t = Wall Thickness,

 σ_f = Nominal Hoop Strength of Slotted Tube, σ = Nominal Hoop Strength of Unslotted Tube,

$$\theta = \frac{\pi}{2} \frac{\sigma_f}{\sigma},$$

$$\lambda^2 = \frac{C^2}{Rt} \sqrt{12(1 - \mu^2)}.$$

TABLE 12.11. Crack Propagation Tests of Heat Treated Zr-2.5 wt% Nb Tubing

Specimen I Identity	Test Temperature	Material Heat Treatment	Condition Hydride	Slot Length, in.	Hoop Strength, ksi	K _C , ⁽¹⁾ ksi $\sqrt{\text{in.}}$	Fracture Mode
18A	23	Not Aged	None	None	126 ^(a)	---	Shear
14A	23	Not Aged	None	None	126	---	Shear
29A	23	Not Aged	None	1/2	92	197	Shear
16A	23	Not Aged	None	1 1/2	33	147	Tension
12A	23	Not Aged	None	2 1/2	28	240	Tension
28A	23	Not Aged	None	4	25	422	Tension
20	23	Aged	None	None	122	---	Shear
29	23	Aged	None	1/2	82	160	Tension
16	23	Aged	None	1 1/2	38	175	Tension
12	23	Aged	None	2 1/2	29	250	Tension
10	23	Aged	None	4	13	215	Tension
19A	23	Aged	~275 ppm	None	100	---	Tension
		Aged	~275 ppm	1/2	39	71	Tension
		Aged	~275 ppm	1 1/2	10.8	57	Tension
		Aged	~275 ppm	2 1/2	4.2	44	Tension
		Aged	~275 ppm	4	16.8	393	Tension
13A	150 °C	Not Aged	None	None	105	---	Shear

(a) This tubular specimen was tested with a Zircaloy-2 liner having a 0.030 in. wall thickness.

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FIGURE 12.15. Comparison of the Crack Propagation Characteristics of Zircaloy-2 and Zr-2.5 wt% Nb Alloys

slots with widths of 10 mils or less and radii ranging 0.050 to 0.125 in. were cut into the OD surface of 2.1 in. ID by 0.17 in. wall tubing to act as stress risers and fatigue crack initiators. Figure 12.16 shows the results of these tests. Tubes leak when the crack on the OD surface reaches about 0.4 in. with a cyclic internal pressurization equivalent to a hoop stress of about 23,000 psi. Brittle fracture occurs when the fatigue crack at the OD surface becomes about 2 1/2 or more times longer than the crack at leakage. There does appear to be some evidence that the critical crack length

FIGURE 12.16. Fatigue Crack Length as a Function of Fatigue Loading Cycles

decreases with increasing amounts of prior fatigue damage. Table 12.12 shows the fracture strength of the Zr-2.5 wt% Nb tubing with a fatigue crack when compared to those with milled slot defects to be appreciably weaker.

Creep and Stress Rupture Properties

To date, relatively little creep data have been obtained from Zr-2.5 wt% Nb tubing under biaxial stress conditions. Tables 12.13⁽²³⁾ and 12.14,⁽²⁷⁾ respectively, present creep data for Zircaloy-2 and Zr-2.5 wt% Nb tubing. The most

TABLE 12.12. *Fatigue Crack Propagation Tests of Heat-Treated Zr-2.5 wt% Nb*

<u>Specimen Identity</u>	<u>Test Temperature</u>	<u>Material Heat Treatment</u>	<u>Condition Hydride</u>	<u>Crack Length, in.</u>	<u>Hoop Strength, ksi</u>	<u>K_C ksi $\sqrt{\text{in.}}$</u>	<u>Fracture Mode</u>
26A	23	Not Aged	None	1 1/2	21	111	---
30-11 SR	23	(a)	None	OD-0.68 ID-0.43	58	148/102	Shear
	23	Aged	None	1.48	21	114	Tension
6	23	Not Aged	None	1.36	23	114	Tension
24 ^(b)	23	Not Aged	(b)	1.32	24	113	Tension
11	23	Aged	None	1.31	23	112	Tension
9	23	Not Aged	None	1.36	23	114	Tension
22	23	Aged	None	1.08	23	84	Tension
27 ^(c)	23	Aged	None	>2	20	153	Tension
22A	23	Aged	None	0.95	23	73	Tension

-
- (a) *This test specimen was in the 30% CW condition and had been subjected to a creep test for about 12,000 hr at 300 °C. Total creep strain was about 2%.*
- (b) *The stress concentrator consisted of a spot weld and local hydriding. Local hydrogen concentration was estimated to be about 100 ppm.*
- (c) *The stress concentrator consisted of an EDM slot with a radius of 0.125 in. located on the ID of the test specimen.*

TABLE 12.13. Stress Rupture Tests of Zircaloy-2 Pressure Tubing

Test	Stress, psi	Temp., °C	Time to Rupture, hr	Primary Creep		Secondary Creep		Third Creep		Secondary Creep Rate, %/hr		Total Strain, %
				hr	% Strain	hr	% Strain	hr	% Strain	min	Average	
N30R-1	57,000	300	4,926	160	0.25	2,500	0.70	2,266	4.02	1.4×10^{-4}	2.8×10^{-4}	4.97
N30R-2	61,700	300	11									
N30R-3	63,500	300	1/6									
N30R-4	59,500	300	788	50	0.22	500	0.42	238	23.7	8.3×10^{-4}	8.3×10^{-4}	24.3
N30R-5	42,000	400	86 1/2									9.2
N30R-6	48,000	371	199	15	0.37	140	1.0	44	11.7	7.1×10^{-3}	7.1×10^{-3}	13.1
N30R-7(a)	57,000	300	209	30	1.1	160	0.85	29	18	5.4×10^{-3}	5.4×10^{-3}	20
N30R-8(a)	54,000	300	1,055	100	0.94	750	0.84	205	19	9.0×10^{-4}	11.0×10^{-4}	21
N30Run-1(b)	56,500	300	861	40	0.25	660	0.54	161	4.7	8.2×10^{-4}	8.2×10^{-4}	5.5
N35R-1	63,500	300	340	18	0.5	60	0.14	246	?		2.2×10^{-3}	?
N35R-2	61,000	300	2,448	100	0.45	1,000	0.65	1,348	3.7	3.2×10^{-4}	6.7×10^{-4}	4.8
N35R-3	57,000	300	18,065 and Continuing	20	0.16					1.9×10^{-5}	3.2×10^{-5}	
N35R-4	64,000	300	7									17
N35R-5	63,000	300	1,696	18	0.85	900	0.80	778	4.85	7.6×10^{-4}	8.9×10^{-4}	6.5
N35R-6	63,500	300	33									12
N35R-7	57,500	350	14 3/4									3.1
N35R-8	57,500	343	210	15	0.40	95	0.85	100	2.60	9×10^{-3}	9×10^{-3}	3.85
N35R-9	49,000	400	16 1/2									7
N35R-11	42,000	400	112									10
N35R-12	50,000	371	70 1/2									11.5
N18R-3	52,800	300	8,000+ Failed Over-Temp.	40	0.12	Not Completed				6×10^{-5}		
N18R-4	58,000	300	50									
N18R-5	56,800	300	1,705	40	0.26	1,300	0.46	365	4.9	3.0×10^{-4}	3.5×10^{-4}	5.6
N18R-6	58,000	300	53									23
K30R-3(a)	61,000	300	17									12
K30R-4(a)	56,600	300	1,174	30	0.85	700	1.13	444	2.3	1.3×10^{-3}	1.6×10^{-3}	4.3
K30R-6	56,600	300	3,024	20	0.17	2,400	1.7	604	0.7	4.5×10^{-4}	6.8×10^{-4}	2.6

(a) Unautoclaved

(b) Hydrided, 275 ppm

TABLE 12.14. *Tubular Stress Rupture Tests, Zr-2.5 wt% Nb Alloy*

<u>Specimen Identity</u>	<u>Test Temperature</u>	<u>Hoop Stress, ksi</u>	<u>Test Duration</u>	<u>Total Strain, %</u>	<u>Average Secondary Creep Rate, % 1 hr</u>
30-2-SR	300 °C	61	1,453 (failed)	>2.4	8×10^{-4}
30-3-SR	300 °C	36	10,725 (disc.)	1.3	5×10^{-6}
	400 °C	25	660 (failed)	3.7 to 10.0	5×10^{-3}
30-11-SR	300 °C	50	11,984 (disc.)	2.1	4×10^{-4}
HT-1	400 °C	30	3,176 (failed)	1.1 to 10.1	1×10^{-3}
HT-2	400 °C	25	1,473 (failed)	>3.5	5×10^{-4}
HT-3	400 °C	20	424 (in test)	0.20	
HT-7	400 °C	16.5	7,250 (failed)	>3.1	2×10^{-4}
HT-1A	400 °C	40	27 (failed)	6.5 to 7.2	
HT-2A	400 °C	30	295 (failed)	2.7 to 8.3	2×10^{-3}
HT-3A	400 °C	25	742 (failed)	>2.2	3×10^{-4}
HT-4A	400 °C	15	746 (in test)	0.50	

Note: HT-1 - Material was 60% cold-worked, solution heat-treated at 880 °C, water-quenched and aged 24 hr at 500 °C.

HT-1A - Material was solution heat-treated at 880 °C, water-quenched, 15% cold-worked and aged 24 hr at 500 °C.

significant thing to be noted is that the creep strength of Zircaloy-2 tubing is appreciably greater under biaxial stress than under uniaxial stress conditions. It should be pointed out, however, that strain data in Tables 12.13 and 12.14 are obtained when the tubular test piece has been cooled and all stresses removed. Uniaxial data are obtained while the test piece is at temperature and under load. Nevertheless, comparing the time to rupture for various stresses, it is apparent that the creep strength of tubing subjected to internal pressures is much greater than a tensile coupon under uniaxial stress loads. (For uniaxial creep data, the reader is referred to Section 6 and Reference 8.)

Most of the tests whose data are tabulated in Table 12.13⁽²³⁾ were run at hoop stresses which were 85 to 98% of ultimate burst strength.

Using the Larson-Miller equation:

$$T(20 + \log t) = \text{Constant}$$

where T = test temperature in $^{\circ}\text{R}$

t = time in hours

c = constant for a given stress

Aungst⁽²³⁾ calculated the equivalent 300 $^{\circ}\text{C}$ rupture times of tests run at temperatures above 300 $^{\circ}\text{C}$ and concluded that Zircaloy-2 tubing had a 20 year life at a stress of 46 ksi. If one applies the same Larson-Miller equation to the Zr-2.5 wt% Nb data of Table 12.14,⁽²⁷⁾ a 20 year rupture stress of about 30 ksi is obtained. This value is less than calculated by Aungst for Zircaloy-2. If one compares the one test 30-2-SR at 61 ksi, the rupture times are comparable. The short time tensile and burst tests at elevated temperatures show the Zr-2.5 wt% Nb alloy to be stronger. Hence, a lower creep rupture strength for the Zr-Nb alloy would not be anticipated

unless appreciable overaging occurs under stress for appreciable time periods at 400 °C. Based on the extrapolations made with the creep data for both the Zircaloy-2 and Zr-2.5 wt% Nb tubing, it appears that further investigation of metallurgical stability of the Zr-2.5 wt% Nb alloy at 300 to 400 °C is warranted. Further creep testing at 300 °C is also needed.

Hydride Effect on Mechanical Properties of Tubes

As yet, there have been no extensive investigations of the effects of hydriding on the mechanical properties of Zr-2.5 wt% Nb alloy tubing. Table 12.10⁽²⁴⁾ shows the effects of hydriding on the "slotted" tube strength and crack propagation behavior of pressure tubing. Certainly at room temperature, tubing is more susceptible to brittle fracture. In general, however, strength does not appear to be grossly impaired. Further investigation of the effects of hydriding on fatigue crack initiation, growth and critical crack length for both irradiated and unirradiated materials appears to be warranted.

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