

# Assessment of Aging of Zr-2.5Nb Pressure Tubes for Use in Heavy Water Reactor

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## ABSTRACT

The pressure tubes of pressurized heavy water reactor (PHWR) operate under high temperature high pressure aqueous environment and are subjected to fast neutron irradiation. In order to assure the structural integrity of pressure tube during service, it is periodically examined by non-destructive examination (NDE) techniques. A typical Pressurized Heavy Water Reactor (PHWR) consists of few hundred horizontally placed coolant channels. The coolant channel comprises of a Zr-2.5% Nb pressure tube encircled by a Zircaloy-4 calandria tube and four garter spring spacers (Zr-2.5Nb-0.5Cu), which prevents these two tubes to come in contact during their service life. The pressure tube carries the nuclear fuel, high temperature high pressure heavy water coolant and is subjected to fast neutron irradiation. The integrity of pressure tube is central to the safety of PHWRs. To ensure this, they are periodically subjected to in-service inspection by nondestructive examination (NDE) techniques. In this paper detailed study has been done on the operating conditions which lead to degradations in the pressure tube with respect to dimensional changes as a result of irradiation creep and growth, deterioration in mechanical properties due to irradiation embrittlement, initiation and growth of new flaws like fretting damage due to debris and fuel element bearing pads. The absorbed hydrogen can also limit the life of a pressure tube due to the degradation mechanisms such as delayed hydride cracking (DHC), hydride blister formation and cracking and hydride embrittlement. It is also one of the regulatory requirement to periodically subject pressure tubes to in-service inspection by employing non-destructive examination techniques. All these aspects have been duly addressed in this paper.

**Keywords:** pressure tube, zircalloy-4, irradiation damage, delayed hydride cracking

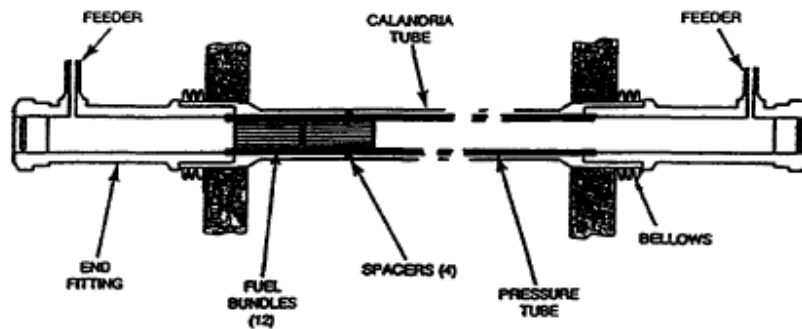
## 1. INTRODUCTION

Zirconium has two crystallographic structures,  $\alpha$ -phase (hexagonal close packed structure) and a  $\beta$ -phase (body centered cubic). To improve the properties of zirconium it is alloyed with tin (Sn), iron (Fe), chromium (Cr), nickel (Ni) and in CANDU reactors with niobium (Nb). In light water cooled nuclear reactors Zircaloy-2 and Zircaloy-4 are the two most frequently used alloys. Two methods are adopted for Zr 2.5 % Nb alloy pressure tube manufacturing for CANDU reactors:

- Heat treatment, solution treatment and aging route.
- Quadruple vacuum arc melted, cold worked using pilger mill route

The design of CANDU fuel channels has evolved to accommodate higher power outputs, reaching a maximum of about 6.5 MW in the current generation of fuel channels. Temperatures and pressures have also increased during this evolution. This has necessitated increases in the length, diameter, and strength of the pressure tubes. As the high pressure/temperature primary coolant is slightly alkaline, the pressure tubes must be adequately resistant to corrosion, as well as to creep and growth. Since CANDU reactors use natural uranium fuel, it is important to use low neutron absorption materials like zirconium alloys in the reactor core. In addition, it is necessary to pay special attention to the design and manufacture of in-core structures to ensure that no material is used beyond that necessary to satisfy the appropriate design code.

Each end of a pressure tube is roll expanded into the hub of a stainless steel end fitting to form a pressure tight, high strength joint. These end fittings provide a flow path for the primary coolant between the pressure tube and the rest of the CANDU primary heat transport system (PHTS) by having a bolted connection to a carbon steel "feeder pipe" whose diameter is about 7 cm. A typical fuel channel design is shown in Figure 1.



**Figure 1: Schematic illustration of one of the fuel channels located in a CANDU reactor core**

The pressure tube acts as a horizontal beam supported at each end by its attachment to end fittings and at intermediate points by the surrounding calandria tube, via the "garter spring" spacers. The inside diameter of the pressure tube is derived from fuel passage and thermo hydraulic considerations. A minimum allowable pressure tube wall thickness is determined by a stress analysis, and then a 0.2 mm allowance for corrosion and wear is added to determine the minimum allowable wall thickness for pressure tube fabrication [1].

The pressure tube design analyses take account of the effects of creep and growth, which causes pressure tubes to permanently increase in length and diameter, and to sag from the weight of the fuel and coolant contained in them. These dimensional changes are illustrated in Figure 6. Pressure tube stresses are evaluated for both beginning of life and end-of-life conditions to account for the pressure tube dimensional changes that occur during their 30 year design life. At the beginning of life, the initial wall thickness and unirradiated material properties apply. At the end-of-life, pressure tube diameter and length have increased, while wall thickness has decreased and material strength has increased. For the end-of-life condition, credit is taken for only a small fraction of the strength increase obtained from irradiation, which occurs mostly in the first few years of operation.

## **2. DESIGN BASIS FOR THE FUEL CHANNEL AND PRESSURE TUBE**

The pressure boundary components for the reactor primary coolant, and the internals of the fuel channel, are designed to withstand the primary coolant flow, temperature, and pressure, including the transient service conditions imposed by the primary heat transport system (PHTS). The design life for all fuel channels installed during initial reactor construction has been 30 years at 80% capacity. The materials of all channel components must exhibit acceptable corrosion resistance and change in properties, etc., during their design life. In addition, the fuel channel must satisfy all of the safety, performance, etc., requirements given below:

The fuel channel components are designed, fabricated and installed to ensure reliable and essentially maintenance-free operation, and a high level of confidence that no channel failure is expected during their design life. It is anticipated that all of the Zr-2.5%Nb pressure tubes in a CANDU reactor core will be replaced after about 30 years operation so that the life of the reactor core can be extended. The replacement of fuel channels is made as efficient and simple as possible to minimize the cost and radiation exposure associated with such retubing of a reactor core.

In-service inspection of fuel channels is performed in accordance with the requirements of CSA Standard N285.4 on the Periodic Inspection of CANDU Nuclear Power Plant Components. Besides mandatory periodic inspections, utilities have instituted additional in-service inspections for reasons of maintenance, operational reliability, to provide assurance to regulators, etc. As much as possible, fuel channel inspections are conducted when a reactor is shut down for other reasons, so these inspections have the minimum possible effect on reactor capacity factors.

To provide a LBB behavior for pressure tubes, the gas annuli between the outside of each pressure tube and the inside of the surrounding calandria tube must be capable of being monitored to detect a leak from a postulated crack in the pressure tube. The leak detection capability provided by the annulus gas system (AGS) of CANDU reactors is

sufficiently sensitive so that the reactor can be shut down and depressurized long before a postulated crack, growing by the delayed hydride cracking (DHC) mechanism, reaches its unstable length.

The material specifications for fuel channel components are listed in the Canadian Standards Association (CSA) Standard N285.6. These define the requirements for impurities, heat treatment, mechanical properties, cleanliness, and testing [2].

### 3. EVOLUTION OF FUEL CHANNEL DESIGN AND MANUFACTURING

Since the early reactors were designed before the irradiation enhancement of pressure tube deformation was known, they did not have adequate allowances for the deformation that occurs during 30 years of operation. The channels associated with these reactors have required an ongoing program of axial repositioning (TUBE SHIFT) and it is expected that many of these channels will be replaced before they have operated for 30 years. The channel designs for later reactors include modifications (longer fuel channel bearings, smaller diameter coils for the annulus spacers and additional spacers associated with each pressure tube) to provide deformation allowances that accommodate a 30 year pressure tube design life. Because all crack propagation, and most crack initiation, for CANDU pressure tubes has been associated with delayed hydride cracking (DHC), most design modifications associated with these tubes have been to eliminate the possibility of DHC occurrence (by reducing hydrogen/deuterium levels or large stress concentrations in the tubes, etc.) and to increase the probability that leak before break (LBB) will be the consequence if DHC does ever occur at operating temperature.

An accelerated corrosion and rate of deuterium (D) pickup from the PHTS coolant has been observed for Zircaloy-2 pressure tubes. As this causes their hydrogen (H) plus D concentration to significantly exceed the terminal solid solubility (TSS) before they have operated for 30 years, all CANDU reactors built after Pickering units 1 and 2 have used Zr-2.5%Nb pressure tubes instead of Zircaloy-2 tubes. Examination of several Zr-2.5%Nb pressure tubes has shown that they do not exhibit the accelerated D pickup rate of Zircaloy-2.

### 4. DEGRADATION MECHANISMS AND AGEING CONCERNS FOR PRESSURE TUBES

Pressure tubes are exposed to temperatures up to 313°C, internal pressure up to 11 M Pa, neutron fluxes up to  $3.7 \times 10^{13} \text{ n/cm}^2/\text{s}$  and fluences up to  $3 \times 10^{22} \text{ n/cm}^2$  (in 30 years of operation at 80% capacity). These conditions cause changes in dimensions and material properties through irradiation damage and micro structural evolution. The tubes are also subjected to corrosion by the slightly alkaline heavy water coolant that flows inside them, with some of the deuterium resulting from this corrosion process being absorbed by the zirconium alloy pressure tubes. Although the overall performance of the more than 10000 pressure tubes installed in CANDU reactors has been good, some of the pressure tubes in early reactors have leaked and two pressure tubes have ruptured, one while at operating conditions, due to delayed hydride cracking (DHC).

The consequence of DHC, when it occurs, usually is pressure tube cracking, leakage, and reactor shutdown for tube replacement. It is expected that leak before break (LBB) will usually be associated with a DHC failure of a pressure tube. However, tube rupture could occur if leakage is not detected before a leaking crack grows to the unstable length. The presence of hydrides has been associated with all crack propagation in zirconium alloy pressure tubes, and most of their crack initiation processes, therefore the H content in new tubes and the D picked up during service need to be minimized.

Analysis of CANDU pressure tubes shortly after being fabricated indicates that they contain a small amount of H. For tubes fabricated before 1990, 5-15 ppm of H is the typical range for the H concentration of the as-fabricated tubes (up to 25 ppm of H was allowed). However, recent improvements in the manufacturing process for Zr-2.5%Nb pressure tubes have significantly reduced the H concentration of tubes after manufacturing, so tubes are now fabricated with an H content of less than 5 ppm. With a typical D ingress rate of 1 ppm (Heq) per year, the consequence of this can be to increase the time it takes to form hydrides in operating pressure tubes by up to 20 years [3].

A few tubes in early CANDU units have had a significant tensile stress concentration because they contained a sharp manufacturing defect. A subsequent review of the pressure tube manufacturing process concluded that top of ingot tubes produced from ingots that contained deep shrinkage cavities may contain lamination flaws.

The manufacturing process has been changed to minimize the size of the shrinkage cavities in ingots and the tops of ingots are now cropped to remove them. Final tube inspection by new techniques with a very high probability of detecting such laminations significantly lowers the risk of having such defects in tubes now entering service.

## 5. PRESSURE TUBE IRRADIATION ENHANCED DEFORMATION

The hexagonal close packed crystal structure of zirconium results in anisotropic deformation and in neutron irradiation causing both irradiation growth (shape change at constant volume in the absence of an applied stress) and irradiation creep (stress dependent constant volume shape change). The deformation of operating pressure tubes (sag, diameter increase, elongation and wall thickness decrease) is predicted to be due to the addition of strains associated with thermal creep, irradiation creep and irradiation growth with the strain rate for each of these being a function of temperature, flux, dislocation density, stress, etc. The design of fuel channels associated with the initial construction of all CANDU and Indian reactors includes allowances for the amount of pressure tube deformation expected to occur during a 30 year operating life.

It is known that irradiation increases tube hardness, yield and tensile strengths, and reduces ductility and fracture toughness. In addition, the susceptibility of Zr-2.5%Nb to DHC increases slightly, and the velocity of DHC increases somewhat, particularly at the inlet end due to its lower irradiation temperature. The consequences of such changes is that pressure tubes become more susceptible to fracture, that is, the margins associated with demonstrating their LBB behavior are decreased and hence the possibility of tube rupture is increased. Changes in tube properties must not result in an unacceptable safety margin on tube rupture.

Since the effect of irradiation on the material properties of Zr-2.5%Nb pressure tubes occurs during the first few years of operation, the values of pressure tube properties after 30 years of irradiation are expected to be adequate to allow Zr-2.5%Nb tubes to achieve their 30 year design life. Test data from the lead operating CANDU reactors, and material testing to high fluences in fast flux facilities of test reactors, will confirm whether or not fracture toughness and DHC velocity values, etc., remain at their saturated values. In addition, pressure tube Fitness-for-Service Guidelines are being developed to provide criteria and assessment methodologies for addressing all aspects related to pressure tube flaw tolerance and how LBB provides a defense in depth limit for the consequences of any pressure tube DHC that may occur.

New pressure tubes have a very large margin between their leaking and breaking conditions. However, as they are operated this margin decreases due to irradiation and D ingress. As a pressure tube's operating life increases, there is an increasing concern that a sufficiently large flaw could exist to precipitate hydrides and initiate DHC. In order for tubes to continue operating, it is required that assessments be done to show there continues to be a high confidence both that DHC will not occur, and also that if DHC does occur the result would be a leak that is detected and the reactor safely shut down before this cracking becomes unstable. To ensure this LBB requirement is satisfied, there must be a high confidence level that the time required for a crack to grow to an unstable length is less than the time available to take appropriate action after leakage is detected. The annulus gas system of CANDU reactors provides a very sensitive detection system for pressure tube leaks, which allows this LBB requirement to be satisfied throughout the 30 year design life for pressure tubes. A corollary of this LBB requirement is that conditions which do not satisfy LBB, such as having a very high H/D concentration in a portion of a pressure tube, must not occur. Therefore, pressure and calandria tube contact must not be allowed to exist long enough for a hydride blister to form and grow at the cold spot of the pressure tube. Since the D ingress rate for Zircaloy-2 pressure tubes is higher than that for Zr-2.5%Nb tubes, the margin on demonstrating LBB for Zircaloy-2 tubes becomes unacceptably small and requires that they be replaced before they have operated for 30 years.

Although degradation of the properties for Zr-2.5%Nb pressure tubes has not been responsible for any operating problems, an improvement in these properties is a highly desirable way to reduce pressure tube rupture concerns. If a crack grows undetected, it will become unstable when it reaches the critical crack length (CCL), which is controlled by the fracture toughness of the material containing the crack. As-fabricated pressure tubes are very tough, but irradiation damages the crystal lattice and changes the tube mechanical properties. There is an initial rapid decrease in toughness to about half its unirradiated value with very little subsequent change.

The toughness values for irradiated pressure tubes cover a very broad range, with some tubes still having quite a high toughness after 18 years of service. Also, the tubes made most recently are tougher than earlier tubes, even though their nominal specification is identical. These observations led to studies to look for subtle micro structural or micro chemical differences that may have caused these differences in toughness.

It was recently observed that tubes containing low concentrations of chlorine have high fracture toughness. Thus the range of fracture toughness is partly related to trace amounts of chlorine, which is a residue of the process used to refine zirconium. The fabrication records revealed that the very tough Zr-2.5%Nb tubes were made from 100% recycled material that had been melted four times. Such quadruple melting of ingots reduces the chlorine concentration to small values and consistently produces tough tubes.

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## 6. ASSESSMENT METHODS AND FITNESS-FOR-SERVICE GUIDELINES

The general requirement for assessments that ensure the continued reliable operation of the pressure tubes in CANDU reactors is to show that:

- Their deformation does not exceed allowable limits (using data primarily obtained by periodic measurements of tube deformation and models developed for predicting this deformation),
- Their service induced deterioration does not exceed allowable limits,
- Their material remains in an adequately tough condition (using data primarily obtained by periodically removing and testing surveillance tubes), and
- There are no mechanisms which could cause significant crack growth.

New pressure tubes have a large margin between the initiation of leaking and unstable rupture. However, as they are operated this margin decreases due to irradiation and deuterium (D) ingress. As a pressure tube's operating life increases, there is an increasing concern that a sufficiently large flaw could cause precipitation of hydrides and initiate DHC. In order for tubes to continue operating, it is required that assessments be done to show there continues to be a high confidence both that DHC will not occur, and that pressure tubes will exhibit LBB, if DHC does occur [4].

Fitness-for-Service Guidelines are being developed within the Canadian nuclear industry to provide acceptance criteria for flaws found in pressure tubes, and to define a standardized approach for assessing the integrity of operating zirconium alloy pressure tubes. The main objective of the assessment process is to ensure that the probability of tube failure remains acceptably low throughout the operating life of the tubes. The Guidelines complement the rules of Section XI of the ASME Code and the requirements of CSA-N285.4 on the periodic inspection of pressure tubes in operating CANDU nuclear reactors. The Guidelines provide the methodology for the evaluation of manufacturing flaws, in-service generated flaws, hydride blisters, and in-service degradation of zirconium alloy pressure tube properties. Trial application of the draft Guidelines has enabled the key pressure tube integrity issues to be more clearly defined and has caused the pressure tube research programmes to be more closely focused on these issues.

## 7. MITIGATION METHODS FOR TO MANAGE PRESSURE TUBE AGEING

Construction debris, such as metal turnings left in the primary heat transport system, can get trapped in the fuel and cause pressure tube fretting damage during the first year or two of operation. The amount of debris that could lead to fretting damage can be minimized during construction by a high level of cleanliness, and during commissioning by the use of strainers at the inlet end of channels. To minimize this corrosion and deuterium pickup, the pH of the coolant, as well as its deuterium and oxygen contents are tightly controlled.

With the exception of the earlier reactors, whose design did not anticipate the full impact of the irradiation enhanced pressure tube deformation, it is expected that pressure tube deformation will not limit the life of CANDU and Indian fuel channels. Only the early reactors do not have sufficient fuel channel bearing length to accommodate all of the irradiation enhanced elongation that is expected to occur during their 30 year design life. The fuel channels for the first seven commercial CANDU reactors, and the first seven Indian reactors, have required an ongoing programme of axial repositioning (TUBESHIFT) to ensure they remain supported by their bearings as they elongate. The channels for the six earliest commercial CANDU reactors are expected to be replaced before they have operated for their

entire 30 year design life. The older reactors have already had all of their fuel channels replaced which has extended the life of these reactor cores 10 to 15 years beyond their initial 30 year design life.

## 8. CONCLUSION

A survey of the performance of CANDU reactors suggest that the measured axial DHC velocities are within 95 % confidence intervals for cold worked Zr 2.5 % Nb material. The critical stress intensity factors measured in pressure tube agrees with the results from small specimen testing results. This confirms critical stress intensity factor that are only very slightly greater than that given by the lower bound curve for cold worked Zr 2.5 % Nb material for same conditions. Therefore the LBB assessment performed in early CANDU reactors remains valid.

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