Application of Leak Before Break Assessment for Pressure Tube in Case of Delayed Hydride Cracking

Gintautas Dundulis\textsuperscript{a}, Remigijus Janulionis\textsuperscript{a}, Albertas Grybenas\textsuperscript{b}, and Vidas Makarevicius\textsuperscript{b}

\textsuperscript{a}Laboratory of Nuclear Installation Safety, Lithuanian Energy Institute, Kaunas, Lithuania, e-mail: gintas@mail.lei.lt

\textsuperscript{b}Laboratory of Material Research & Testing, Lithuanian Energy Institute, Kaunas, Lithuania

Keywords: Leak before break, Delayed hydride cracking, Fracture mechanic, R6 method, J integral.

1 ABSTRACT

Ignalina NPP contains RBMK-1500 type reactor. RBMK reactor is graphite-moderated with a water-cooled reactor core. The fuel cell assembly is located in the centre of the moderator column and consists of a pressure tube into which the fuel element assembly is inserted and through which the coolant flows. As a constructional material for manufacturing of pressure tubes zirconium alloys Zr + 2.5 \% Nb are used. The hydrogen absorbed by zirconium alloy during corrosion process is one of the factors determining lifetime of pressure tube. When hydrogen concentration in pressure tube exceeds solubility limit initiation and development of delayed hydride cracking (DHC) can take place. The formation of hydrides under certain conditions can reduce resistance to brittle fracture. In this work the evaluation of the influence of hydrides on the fracture of pressure tube and application of leak before break (LBB) for these tubes with DHC was performed. The deterministic analysis of the pressure tube employing LBB concept was carried out using experimental data. Performed deterministic LBB analysis confirms that the pressure tube comply with LBB requirements.

2 INTRODUCTION

The Leak-Before-Break (LBB) concept is associated with the nuclear power plant design principles as regards pipe failures and their safety implications. During the past years, LBB has received increasing applications as a criterion for assessing or upgrading the safety of existing plants. LBB is complex analysis showing that if the started surface crack will grow to the through-wall crack, through this crack some amount of coolant will flow; this leak during definite time can be detected by leak monitoring system and through-wall crack will remain stable under all predictable loading conditions (Wilkowski, 2000). To ensure the LBB concept, it is required that timely detection, confirmation of leak and appropriate operator action to shut the reactor down to a cold depressurized state can be done before the growing crack exceeds the critical crack length (Cheadle et all, 1996, Puls et all, 1998, Serpan et all, 1997). The LBB case of pressure vessels and piping is achieved with the sufficient margin between detectable leak and catastrophic break. Therefore, the respective characteristic crack sizes, i.e. leak size (the crack size at which leakage amount is great enough to be detected) and the break size (the crack size at which crack propagation becomes unstable) are two important governing parameters for the LBB case (Xie, 1999a). The evaluation of crack opening area, investigation of the effects of crack morphology parameters on leak rate is important in LBB analysis. The evaluation of the crack opening area (displacement) plays a nodal role in the demonstration of leakage size crack corresponding to the detectable leak flow rate from the flawed piping system. The linear elastic and elasto-plastic models are used for crack opening area analysis (Ghosh et all, 2009, Kim, 2008). The crack morphology is an important parameter in the calculation of the leakage flaw size. The analysis results of influence the crack morphology illustrate the importance of defining the surface roughness and number of turns in conducting leak-rate analyses (Rudland et all, 2002). Different selections of these input parameters can significantly affect the LBB results.

The LBB concept is employed as defence in depth in pressure tube reactors in order to avoid an unstable failure in pressure tubes (Xie, 1999b, Puls, 1998, Park, 2002, Cheadle, 1996, Park, 1999, Dundulis, 2006). Two types of pressure tube reactors are under operation in Europe: the CANDU6 reactor in Romania, with heavy water as coolant and moderator, equipped with horizontal pressure tubes and the RMBK-1500 in
Lithuania water cooled, graphite moderated vertical pressure tube reactor. As opposed to the single pressure vessel PWR concept, these reactor types are based on a number of individual pressure vessels, called pressure tubes (PT).

The application of LBB methodology for Ignalina NPP pressure tube manufactured from TMT-2 Zr-2.5 Nb alloy is presented in this paper. Zirconium alloys pick up hydrogen during operation as a consequence of corrosion reaction with water and delayed hydride cracking (DHC) failures may occur. The DHC velocity, influence of irradiation and hydrides to the mechanical properties and fracture parameters were evaluated in this analysis. As is mentioned before, the roughness of the crack surface can significantly affect the LBB results, for that reason the irregularities of the crack surface was measured and evaluated in the calculation of leak flow rate through a DHC crack.

The approach presented in the paper is designed to demonstrate LBB applicability to RBMK-1500 pressure tube. The performed analysis using experimental data show that LBB concept is possible to employ as defence in depth of pressure tube reactors.

3 DATA FOR LBB ANALYSIS

3.1 Mechanical properties and fracture parameters

The pressure tubes are made from zirconium alloys (Zr+2.5% NB). The experimental investigations were performed of pressure tubes, which are manufactured using the heat treatment technology TMT-2. The TMT-2 treatment includes inert gas (He+Ar) cooling after the tube has been heated to a temperature in the \((\alpha Zr + \beta Zr)\) phase in the upper part of Zr-Nb alloy diagram, before the last cold rolling. Further cold rolling is done to a 15-25% reduction in area and followed by ageing in the \(\alpha\)-phase (24 hours at temperature 530-545 \(^{0}\)C).

The mechanical properties of the pressure tube are changing at the influence of the radiation. Compared with initial properties, the yield limit is increased to 32-57\%, ultimate strength to 26-49\%, the relative elongation is decreased 1,5-3 times (Ignalina, 2002, Coleman C. et al, 2007). The test results confirm the known trend of material strength properties increase and plasticity decrease under neutron radiation. Stabilization of short-term mechanical properties of PT tube material occurs approximately after the fluence \((1-1.5) \times 10^{24} \text{n/m}^2(\text{E>1 MeV})\).

The experimental investigation of the influence of the hydrogen on mechanical properties and fracture parameters was performed. The dependence of the yield strength and strength limit of Zr-Nb alloy on temperature and hydrogen concentration is presented in Figure 1. The dependence of the J-integral unirradiated Zr-Nb alloy on hydrogen concentration is presented in Figure 2. Obtained data show (Fig. 1) that compared to the irradiated PT material, the source PT TMT-2 material has the lower values of mechanical properties however the temperature effect practically has no influence: with the increase of temperature mechanical properties decreases for both nonirradiated and irradiated material. In the case of hydrided material with an increase of temperature influence of hydride phase decreases and at the temperature above 170 \(^{0}\) becomes little noticeable. It is assumed that influence of hydrogen on the mechanical properties of the irradiated material at the same concentrations of hydrogen and in temperature range 170 - 300 \(^{0}\), as in the case source material, is insignificant. That is related to the gradual decrease of the volume of hydride phase in the zirconium alloy because it dissolves at higher temperatures.

The dependences of \(J_{0.2}\) - integral for nonirradiated Zr 2.5 Nb alloy on hydrogen concentration at temperatures 20, 170 and 300 \(^{0}\) are presented in Fig. 3. Obtained experimental data can be sufficiently well described by the equation:

\[
J_{0.2(H)} = J_{0.2(T)} e^{kC},
\]

where \(J_{0.2(T)}\) is the value of \(J_{0.2}\) - integral for the non-hydried material at a given temperature; \(k\) and \(C\) - calculated coefficient at a given temperature and hydrogen concentration (ppm).

Using an equation (1) and taking into account the temperature of the terminal solid solubility (\(T_{\text{TSSD}}\)) of hydrogen, the generalized curves of the \(J_{0.2}\) dependences on the temperature and hydrogen concentration for the nonirradiated material were built. It was found that the obtained calculated curves of \(J_{0.2}\), taking into
account the temperature of the complete dissolution of hydrides in the alloy, sufficiently well will agree with the experimental data and possess certain conservatism, especially at elevated temperatures.

The curves of temperature dependence of the $J_{0.2}$ on the postulated concentrations of hydrogen for irradiated TMT-2 material, calculated using the (1) are given in Figure 3.

Figure 1. The temperature dependences of the ultimate strength ($R_m$) and yield strength ($R_{p0.2}$) of the initial and hydrided PT Zr 2.5Nb alloy with TMT-2 treatment and data of irradiated PT material. Neutron fluence: $1.2 - 5.3 \times 10^{25}$ n/m$^2$; $3.4 - 6.5 \times 10^{25}$ n/m$^2$. (Coleman, 2007).

Figure 2. The $J$-integral as a function of hydrogen concentration for Zr 2.5Nb TMT-2 material at temperatures 20, 170 and 300 °C. Data of post-reactor studies for irradiated TMT-2 pressure tubes (neutron fluence $6.5 \times 10^{25}$ n/m$^2$) (Coleman, 2007).
Figure 3. The calculated curves of the temperature dependence of $J_{0.2}$ on hydrogen concentration for the irradiated TMT-2 PT material. Curve 1 - experimental data for the irradiated TMT-2 pressure tube (neutron of fluence $6.5 \times 10^{25} \text{n/m}^2$) (Coleman, 2007).

3.2 Investigation of delayed hydride cracking

Testing of the DHC velocity in the RBMK-1500 reactor PT TMT-2 material was carried out. In these experimental investigations the sections of the PT were hydrided to produce required hydrogen concentration using an electrolytic method and diffusion annealing treatment (Lepage, 1998). Predetermined amounts of hydrogen ranging from 27 to 76 ppm were added to the unirradiated sections of the PT by electrolytically depositing a layer of hydride on the surface of the PT material followed by dissolving hydride layer by diffusion annealing at elevated temperature. The compact toughness specimens were machined from hydrided PT according to ASTM E-399 requirements and DHC velocities were measured at the temperatures 200, 250 and 275 °C.

The tested DHC velocity data as a function of 1000/T is plotted in Figure 4.

Figure 4. Dependency of the DHC velocity on temperature for unirradiated RBMK TMT-2 Zr-2.5%Nb alloy.

It is known that the neutron irradiation increases DHC velocity. DHC velocity for the zirconium alloys with fluence $7.7 \times 10^{25} \text{n/m}^2$ can increase up to 50 times (Hosbons, 2000).
Major factor influencing DHC velocity before and after of the irradiation of Zr-2.5 Nb material is changes in the yield strength of the alloy (Oh, 2000). For evaluating the DHC velocity of the irradiated pressure material, the experimentally determined dependence (IAEA-TECDOC, 2004.) of the DHC on Rp0.2 for the pressure tube TMT-2 was used. It was found, that in comparison with the source material, DHC velocity in the irradiated TMT-2 material after the fluence of neutrons 6.5·10²⁵ n·m⁻² increases 25 times. For conducting the calculations the following dependence is obtained:

\[ V_{DHC} = 25*0.000091*\exp(-5.54676*1000/T) \]  

(2)

3.3 Crack surface irregularities

The crack surface irregularities have particular influence for the leak rate through a crack in case of calculation for LBB analysis. To estimate more accurately leak through the postulated flaw, roughness height on the DHC surface was measured from the fractured test specimens. The optical microscope was used for this study. About 95% of all measured irregularities on DHC surface have uniform roughness height, which depends on test temperature. General view of such surface irregularities is shown in Figure 5a. Obtained measurements data show that increase of temperature on DHC surface forms several larger drops in height. These ribs on the fracture surface are oriented to the direction of crack growth. Maximum height of some individual ribs is 100 µm and higher, average height is 86±16 µm at 283ºC (Figure 5b). Average frequency of ribs with height 55-110 µm in one specimen is 2-3. To calculate leak flow rate through a DHC crack we assumed that a height of uniform roughness was 28 µm and repeated rib roughness was 90 µm with frequency 3 in transverse direction.

![Figure 5. LHP fracture surface, test temperature 283°C (a); dependence of average roughness height on DHC testing temperature (b).](image)

4 DETERMINISTIC LBB ANALYSIS BASED ON DHC

The deterministic analysis of compliance to LBB concept of the RBMK PT TMT-2 has been performed according to the requirements of VD-E-98-03 (Guidance, 1998). The minimal allowable wall thickness with possibly the largest diameter of pressure tube was taken for conservatism of a deterministic LBB analysis - tube wall thickness – 3.6 mm and outside diameter – 90.0 mm. The experimental mechanical properties, fracture parameters and DHC velocity of the fuel channel were used for analysis. The analysis was performed at loading of the normal operational conditions (NOC).

4.1 Determination of through-wall crack critical length

Critical length of through-wall crack was determined using the following two methods:

- Method R6 Option 2 Category 1 (International, 1988);

The results obtained using both methods are comparable. However, R6 method gives more conservative results. The results of the analysis are presented in Table 1. The influence of the hydrogen concentration at
corresponding temperature was evaluated using tested material properties and fracture parameters of the zirconium alloys.

**Table 1.** Through-wall crack critical length.

<table>
<thead>
<tr>
<th>Loads</th>
<th>Hydrogen concentration, ppm</th>
<th>Through-wall crack critical length, mm</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>Method R6</td>
</tr>
<tr>
<td>Normal operation conditions</td>
<td>0</td>
<td>72.9</td>
</tr>
<tr>
<td></td>
<td>20</td>
<td>72.9</td>
</tr>
<tr>
<td></td>
<td>70</td>
<td>70.1</td>
</tr>
</tbody>
</table>

4.2 Determination of Crack Opening Function

The function of crack opening is one of the basic initial data for leak rate calculations. Elastic-plastic finite element analysis was performed for construction of a given function. The computer code CASTEM 2000 (Axisa, 1996) was used for this analysis. 3D FE model was prepared for modelling of the function of crack opening. The FE model of PT and the von Misses stress a distribution in the PT is presented in Figure 6. The function is constructed for lower parts of PT using normal operation loading. The results of analysis are presented in Figure 7. As it is visible from presented results, crack opening on outside is approximately 20% larger, than on inside. For critical crack length of 70.1 mm in PT lower part at NOC, the maximum crack opening is about 0.78 mm on inside and about 1.0 mm on outside surface.

![Image of finite element model of pressure tube with through-wall crack](image1)

**Figure 6.** The finite element model of pressure tube with through-wall crack: a- finite element model of PT, b - the von Misses stress a distribution in the PT.

![Diagram of crack opening displacement](image2)

**Figure 7.** The crack opening displacement as a function on postulated crack length under NOC at inside and outside surface.
4.3 Determination of leak rate function

Determination of leak rate is performed for NOC. The crack growth by mechanism of DHC was assumed in the leak rate analysis. The measured irregularities were evaluated in the calculation of leak flow rate through a DHC crack.

For leak rate calculations the computer program SQUIRT v.2.4 was used (Ghadiali, 1996). It was considered, that the crack opening is of elliptical shape, the wall thickness is equal to 3.94 mm, and discharge coefficient is equal to 0.95. The calculations were performed using crack opening function as shown in Figure 7. The required minimum level of the leak rate (equal to 100 kg/h) is reached when length of through-wall crack is about 28 mm (Figure 8). It was found that required minimum level of leak is reached at length of through-wall crack more that two time lower of calculated length of critical through-wall crack, i.e. 28<70.1/2.

![Figure 8](image)

**Figure 8.** The leak rate as a function on postulated crack length under NOC.

4.4 Stability analysis of the postulated cracks

The stability analysis of crack growth is very important in the LBB analysis. In Figure 9, the dependence of fracture parameters on load factor is presented. The presented results show, that if pressure increases 1.4 times (8.84*1.4=12.38 MPa), J-integral reaches 35 kN/m and the crack tip opening displacement reaches 0.04 mm. It is almost 3 times less than critical value of critical J-integral (calculated critical J-integral is 103.5 kN/m). Thus, it is possible to conclude, that unstable growth of the crack of postulated sizes will not occur.

![Figure 9](image)

**Figure 9.** Dependence of fracture parameters on load factor at l_leak.
4.5 Analysis of postulated crack growth

According to the requirements of VD-E-98-03 (Guidance, 1998) leak should be detected less than in 1 hour after shutdown of the reactor. In case of leakage detection in reactor core the reactor shall be shutdown using emergency reactor cooling procedure. According to this procedure the water temperature in the main circulation circuit decreases by 30°C per hour. The pressure also reduces along with temperature. The change in temperature and pressure has influence on critical crack size.

During cooling of the reactor and pressure in PT decreases, the length of critical crack $l_c$ changes. On the one hand, a decrease in the temperature and pressure increases its value, on the other hand, the decrease of temperature leads to the precipitation of the hydride phase, and the length of the hydride crack $l_{DHC}$ gradually increases with the speed depending on the temperature. Since the length of $l_{DHC}$ during the time of reactor cooling increases, there is a probability that at some moment of time its length becomes equal with a length of $l_c$ and will lead to the PT failure.

In order to estimate the possibility of such event for PT TMT-2, dependences of $l_c = f(t)$ and $l_{DHC} = f(t)$ were made. At each moment of time the condition must be satisfied:

$$\Delta l = l_c - l_{DHC} > 0$$  \hspace{1cm} (3)

The prognosis results for the DHC crack growth are presented in Figure 10. The critical crack length in zirconium alloy without hydrogen and with hydrogen concentration of 70 ppm, at decreasing of temperature during shutdown of reactor is presented in this figure too.

The critical crack length has a tendency to increase during all reactor shutdown process. The possible intersection of the growing crack curve with a critical crack size curve could not be reached during shutdown of the reactor. Therefore the time required to detect a leak and to shutdown reactor before rupture of the PT will be sufficient.

![Figure 10. LBB assessment results for reactor emergency shutdown procedure.](image)

5 CONCLUSION

The experimental investigation of influence of the hydrogen concentration up to 140 ppm on the mechanical properties and fracture parameters of the RBMK-1500 PT TMT-2 Zr 2.5Nb alloy was carried out. The results show that strength of the alloy increases and J-integral decreases with the increase of hydrogen concentration in the temperature range from 20 to 170 °C. However, influence of hydrogen becomes insignificant at the higher temperatures.
Temperature dependence of delayed hydride cracking velocity was determined in the temperature range 200 - 275 °C. DHC velocity for the irradiated pressure tube was evaluated using experimentally determined dependence of the DHC on $R_{p0.2}$ for the TMT-2 material.

The deterministic analysis of compliance to LBB concept of the RBMK pressure tubes TMT-2 confirmed, that the length of postulated crack, at which necessary leak rate is reached, is less than half-length of critical through-wall crack. According to the prognosis the DHC crack growth will not reach the critical crack size during shutdown of the reactor. Therefore it is possible to conclude that RBMK PT after 17 years operation fulfils the LBB requirements in case of possible DHC cracking.

Acknowledgement.

The study was supported by Ignalina NPP, Lithuanian Science Foundation and Network of Excellence of Nuclear Plant Life Prediction (NULIFE) within the 6th EU Framework Programme. The authors also want to express gratitude to the administration and technical staff of the Ignalina NPP, for providing information regarding operational procedures and operational data.

REFERENCES


