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ScienceDirect
Journal of Nuclear Materials 362 (2007) 300–308
journal of nuclear materials
www.elsevier.com/locate/jnucmat

The irradiation effects on zirconium alloys

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Abstract

Pressure tube samples were irradiated under helium atmosphere in the TRIGA Steady State Research and Material Test Reactor of the Romanian Institute for Nuclear Research (INR). These samples are made of the Zr–2.5%Nb alloy used as structural material for the CANDU Romanian power reactors. After irradiation, mechanical tests were performed in the Post Irradiation Examination Laboratory (PIEL) to study the influence of irradiation on zirconium alloys mechanical behaviour. The tensile test results were used for structural integrity assessment. Results of the tests are presented. The paper presents, also, pressure tube structural integrity assessment.

PACS: 28.20.Fc; 28.41.Qb; 28.50.Dr; 46.50.+a; 61.80.Lj; 62.20.Fe; 62.20.Mk; 68.35.Gy

1. Introduction

The reactor irradiation of zirconium alloys leads to degradation of their mechanical properties as a result of fast neutron action. Any irradiation induced defect isolated in the lattice, which does not adhere to the dislocation loops, influences macroscopic dimensional parameters, e.g. material form and volume changes. The observation of dimensional changes of the Cernavoda CANDU fuel channels and their pressure tubes as well as the detection of defects induced during normal operation were performed by in-service inspection according to Canadian standards [1,2]. During the last planned outage [3,4], an inspection of Cernavoda NPP Unit 1 was performed for the following purposes:

(a) pressure tube inside diameter measurements,
(b) gap between pressure tube and calandria tube measurements,
(c) tube sagging measurements,
(d) fuel channels elongation,
(e) pressure tubes defects detection and characterization.

In service inspections in some Canadian reactors reveal the following types of flaws after periods of operation [5–7]:

(1) scratches due to fuel bundles,
(2) pitting corrosion,

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doi:10.1016/j.jnucmat.2007.01.160

(3) bearing pad flaws (BPF) due to fuel bundles pads on the pressure tube inside surface,
(4) debris fretting flaws (DFF) due to impurities.

It is important to make an assessment of the pressure tubes structural integrity [8–10]. This evaluation has been performed in our paper by means of the Failure Assessment Diagrams Methods (FAD) in the British standards [11,12].

For this assessment zirconium alloys used as structural material for the CANDU Romanian power reactors were irradiated in the Romanian TRIGA Steady State Research and Material Test Reactor. The irradiation campaign aimed to obtain data on pressure tube behaviour during service in the Cernavoda NPP Unit 1, a CANDU 600 nuclear reactor. Data obtained from this irradiation are to be compared with data obtained from the in service inspection. The data obtained from post-irradiations assessment combined with other laboratory experiments are to be used in pressure tube integrity assessment with the computer codes.

2. Experimental

2.1. Irradiation campaign and instrumentation

Prior to this study, four irradiation campaigns have been performed using the irradiation capsule type C5 in the TRIGA reactor. For this experiment, the irradiation was performed intermittently during the fifth campaign of irradiation between October 11, 1995 and December 18, 2001.

The testing conditions for these pressure tube samples were: helium with temperatures at 530–550 K, and pressure at 0.1–0.6 MPa, neutron flux ($E > 1$ MeV) 1 to 2×10^{13} $cm^{-2}\ s^{-1}$, neutron fluence ($E > 1$ MeV) up to 1.0×10^{20} cm^{-2}. TRIGA reactor power was not constant, due to multiple utilizations.

Capsule type C5 is an irradiation device designed for irradiating reactor structural materials. It was designed and manufactured entirely at the Institute for Nuclear Research at Pitesti, in 1984.

In order to test pressure tube irradiation samples (Zr–2.5%Nb) a new fuel holder was designed and instrumented to give extensive data about the sample's irradiation conditions. The C5 capsule is instrumentally equipped to monitor temperature, neutron flux and pressure.

The operational conditions and limits are a maximum allowed pressure of 6 MPa and a maximum sample temperature of 565 K.

The capsule was designed for the following objectives: the characterization of the mechanical behaviour and the material microstructure evolution during irradiation and improvement of the simulation methods for different phenomena specific to in-core operation of structural materials.

The next campaigns were for zirconium alloy tests, so the capsule was redesigned to test Zr–2.5%Nb samples. The samples were provided with Cr–Al thermocouples to measure temperature and collectrons to measure thermal flux. Thermal flux was correlated by neutron computation with the fast flux. Neutron fluence was evaluated by neutron calculations correlated with gamma spectrometry measurements on the Zr–2.5%Nb irradiated samples. The fast flux was estimated taking into account the capsule position in the reactor core.

2.2. PIEL tests

After the irradiation process the pressure tube samples have to be tested to evaluate the changes in the mechanical properties [13,14]. In the Post Irradiation Examination Laboratory (PIEL) the tensile testing machine is an INSTRON 5569 model. The machine uses the Merlin software to process the data obtained from the mechanical tensile tests. The tests were performed according to the procedures and standards given in references [15,16].

The tests have been performed to record or evaluate the following mechanical characteristics:

- the strain–stress diagrams and load extension (numeric and plotted);
- the Young modulus;
- the yield strengths (offset method at 0.2%);
- the elastic limit and
- the ultimate tensile strength of the samples.

First, an initial test on un-irradiated samples was performed in order to obtain the tensile test machine stiffness. The test was made with the machine furnace to assess the correlation between the required temperature and real probe temperature measured by a thermocouple.

In the PIEL the tensile tests were performed according to ASTM procedures using the INSTRON 5569 testing machine. From the PIEL tensile tests performed on the Zr–2.5%Nb samples the following mechanical characteristics were obtained:

- Real stress–strain diagrams,
- yield strengths (2% offset method),
- ultimate tensile strength (UTS) and
- fracture elongation of the samples were evaluated.

Metallographic examination after the tensile tests was made to analyze the irradiated samples surface macroscopic status and characteristics of the fractured area [17].

The primary data of the tensile tests were used to infer Ramberg–Osgood relationship parameters. The Merlin software was used to process the tensile experimental diagrams.

3. Assessment of pressure tube structural integrity

3.1. Constitutive correlations obtained from the irradiated Zr–2.5%Nb samples

Based on the primary curves processing, the following correlations were obtained that can be used in finite element computer codes.

The yield strength in the longitudinal direction is described by:

$$\sigma_{0.2} = -0.8843 \cdot (T - 273) + 757 \quad (1)$$

and in the transversal direction it becomes

$$\sigma_{0.2} = -0.4068 \cdot (T - 273) + 818.8, \quad (2)$$

with T in K and $\sigma_{0.2\%}$ in MPa.

The (UTS) ultimate tensile strength in the longitudinal direction is given by

$$\mathrm{UTS} = -1.2636 \cdot (T - 273) + 853.7 \quad (3)$$

and in the transversal direction it becomes:

$$\mathrm{UTS} = -1.356 \cdot (T - 273) + 843.8, \quad (4)$$

with T in K and in UTS MPa.

The Ramberg–Osgood equation is used to describe the stress–strain relationship in the yield region of the stress–strain diagram. The material tensile behaviour in the elastic plastic domain is modelled with the Ramberg–Osgood correlation.

$$\frac{\varepsilon}{\varepsilon_0} = \frac{\sigma}{\sigma_0} + \alpha\left(\frac{\sigma}{\sigma_0}\right)^n, \quad (5)$$

where ε_0 and ε are elastic and actual strain respectively, σ_0 and σ are yield strength and actual strength respectively, α and n represents experimental data fitting constants.

Finite Element Analysis (FEA)-Crack is a fracture mechanics computer code that performs strain–stress analyses by means of the Finite Element Method on components with flaws like cracks and extracts the fracture parameters required for performing structural integrity analysis. The paper presents the results for transversal direction irradiated samples.

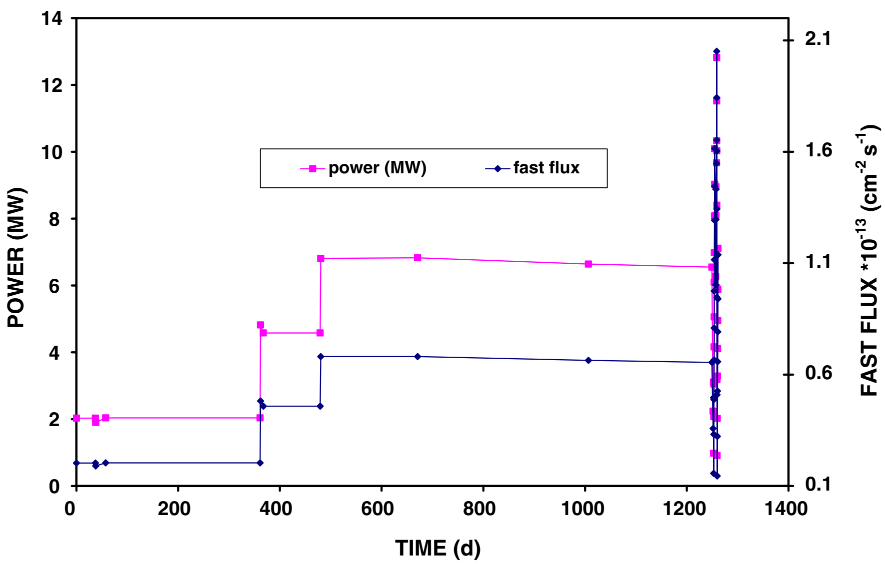

Fig. 1. Capsule 5 monitored fast flux and reactor power for irradiation history in the fifth campaign. Conditions: Zr–2.5%Nb samples in helium with temperatures at 530–550 K, and pressure at 0.1–0.6 MPa, neutron flux ($E > 1$ MeV) 1×10^{13} to 2×10^{13} cm^{-2} s^{-1}, up to a fluence of 10^{20} cm^{-2}.

3.2. Structural integrity assessment of a pressure tube with a sharp flaw

The structural integrity assessment for an irradiated pressure tube with a crack on the inside tube face is performed using Failure Assessment Diagrams (FAD) [18]. In the $L_r < L_{r\max}$ range the failure assessment curve (FAC) is given by the relationship

$$f_1(L_r) = \frac{0.3 + 0.7\exp(-0.6L_r^6)}{\sqrt{1 + 0.5L_r^2}}, \tag{6}$$

where L_r is the limit load parameter (collapsing parameter).

The FAC is useful for material analysis where the yield strength and ultimate tensile strength are known. Finite element analyses in the elastic–plastic domain could be performed knowing the Ramberg–Osgood correlation.

Points of abscissa in FAC are given by collapsing pressures, which are computed in two situations.

First the global collapsing pressure without stresses on the crack faces is given by

$$P_L = \sigma_{0.2}\left(\frac{c}{R_1 M} + \ln\left(\frac{R_2}{R_1 + c}\right)\right), \tag{7}$$

with

$$M = \left(1 + \frac{1.61 \cdot b^2}{R_1 \cdot c}\right), \tag{8}$$

where c is crack depth, $2b$ is the inside surface crack length, R_1 is the pressure tube inside diameter and R_2 is the pressure tube outside diameter. The local collapsing pressure without stresses on the crack faces is given by

$$P_L = \frac{\sigma_{0.2}}{2(s + b)}\left(2s \cdot \ln\left(\frac{R_2}{R_1}\right) + 2b\ln\left(\frac{R_2}{R_1 + c}\right)\right), \tag{9}$$

with

$$s = \frac{bc\left(1 - \frac{c}{w}\right)}{M \cdot R_1\left(\ln\left(\frac{R_2}{R_1}\right) - \ln\left(\frac{R_2}{R_1+c}\right)\right) - c}, \tag{10}$$

where s is a dimensional parameter (m) and w is wall thickness.

These correlations describe collapsing pressure evolution in both cases, and L_r can be plotted as

$$L_r = \frac{P}{P_L}, \tag{11}$$

where P is the internal pressure.

The toughness parameter K_r is inferred as

$$K_r = \frac{K_I}{K_{IC}}, \tag{12}$$

where K_I is the stress intensity factor and K_{IC} is the material toughness in the plane strain state.

4. Results and discussion

In this section the PIEL tests results and structural integrity assessment of the irradiated Zr–2.5%Nb samples are presented.

4.1. Irradiation campaigns

The purpose of this irradiation was to obtain data useful for assessment of pressure tube behaviour

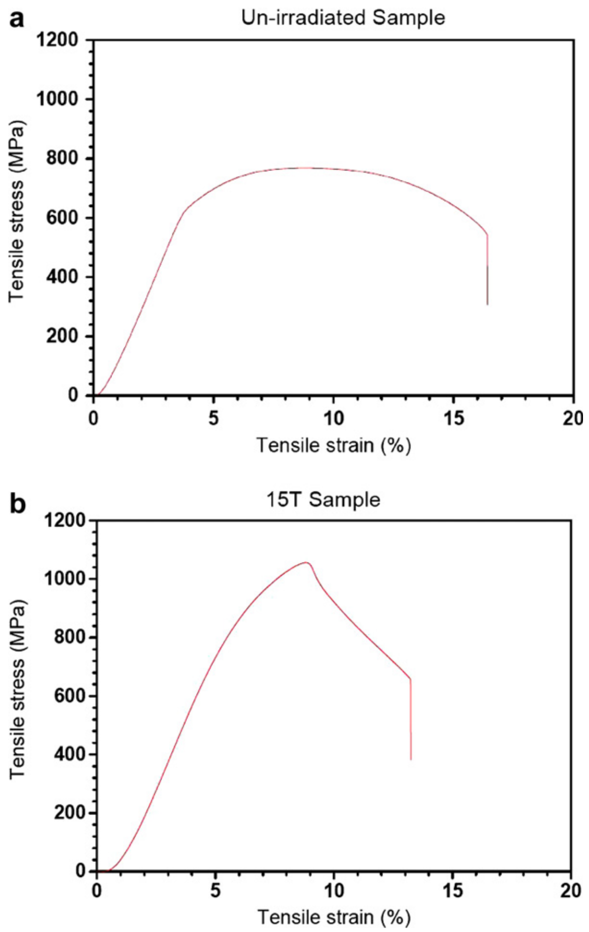

Fig. 2. Strain–stress curve for an unirradiated sample (a), and for the irradiated sample T15 (b). Conditions: deformation rate: $0.1\ \text{min}^{-1}$, test temperature 343 K, sample geometry, ASTM E8M Plate Type sample, Gage length 25 mm; Width 6.5 mm; Thickness 4.2 mm; no heat treatment.

during operation in the Cernavoda Nuclear Power Plant (NPP).

In Fig. 1 the C5 capsule irradiation history for the fifth campaign is presented. Two thermal neutron flux transducers records together with the TRIGA reactor power record are presented.

The Zr–2.5%Nb pressure tube tensile samples were: four irradiated samples from the pressure tube transversal sections 11–14L, four irradiated samples from the pressure tube transversal rolled areas 15T to 18T and eight irradiated samples from the pressure tube transversal rolled areas 1–8TM.

4.2. PIEL test results

Merlin software was used to process data obtained by the INSTRON 5596 test machine. The sample geometry is according to ASTM E8M [15]. A strain–stress curve for an un-irradiated sample of Zr–2.5%Nb at 343 K is presented in Fig. 2(a). It may be compared with the strain–stress curve for the irradiated transversal sample 15T at 343 K presented in Fig. 2(b). In the elastic region the sample behaviours are similar, but differences are evident in the plastic region. UTS increases with irradiation as can be seen in the Table 1. Strain decreases with irradiation from a comparison of Fig. 2(a) and (b).

The micrography of the crack front propagation is presented in Fig. 3. The initiation of a 3.5 mm long crack in the material is noticed. This has been propagated from the fracture surface external side of the sample towards its internal part.

The tensile characteristics and the temperatures in the INSTRON 5569 furnace for the Zr–2.5%Nb irradiated samples and the processed data are presented in Table 1. Mechanical characteristics of the irradiated samples: elastic limit engineering and real values, yield stress engineering and real values, and ultimate tensile strength (UTS) engineering and real values at different temperatures are presented.

4.3. Assessment of the PIEL data

In Table 2 the Ramberg–Osgood coefficients: Young's modulus E, and experimental fitting constants α and n, are presented for different temperatures for each irradiated pressure tube sample. In Table 3 the Ramberg–Osgood correlation coefficients and mechanical characteristics elastic limit, yield stress and UTS at different temperatures of the irradiated samples, are presented based on the

Table 1
Tensile parameters characteristics for the Zr–2.5%Nb samples irradiated with a neutron flux of 3.5×10^{20} cm^{-2} ($E > 1$ MeV)

Sample	Temperature (K)	Strengths (MPa)						Remarks
		$\sigma_{0\,eng}$ [a]	$\sigma_{0.2\,eng}$ [b]	UTS_{eng} [c]	$\sigma_{0\,real}$ [d]	$\sigma_{0.2\,real}$ [e]	UTS_{real} [f]	
Unirradiated	**343**		**642**	**768**				
11L				**944**			**1114**	failed
15T		**657**	**800**	**1056**	**653**	**901**	**1138**	
1TM		**800**	**942**	**1003**	**887**	**1044**	**1066**	
5TM		**875**	**975**	**1017**	**892**	**1009**	**1078**	
16T		**725**	**833**	**933**	**808**	**867**	**983**	
2TM		**800**	**900**	**927**	**842**	**944**	**978**	
6TM		**753**	**833**	**868**	**820**	**932**	**924**	
13L	**523**	**660**	**750**	**806**	**725**	**778**	**864**	
17T		**717**	**817**	**832**	**760**	**835**	**871**	
3TM		**800**	**867**	**884**	**853**	**918**	**928**	
7TM		**800**	**887**	**892**	**833**	**929**	**939**	
14L	**573**	**533**	**687**	**769**	**667**	**732**	**824**	
18T		**653**	**750**	**784**	**700**	**803**	**820**	
4TM		**740**	**833**	**835**	**800**	**874**	**874**	
8TM		760	780	773	800	808	808	

[a] $\sigma_{0\,eng}$ – elastic limit, engineering values.
[b] $\sigma_{0.2\,eng}$ – yield strength, engineering values.
[c] UTS_{eng} – ultimate tensile strength, engineering values.
[d] $\sigma_{0\,real}$ – elastic limit, real values.
[e] $\sigma_{0.2\,real}$ – yield strength, real values.
[f] UTS_{real} – ultimate tensile strength, real values.

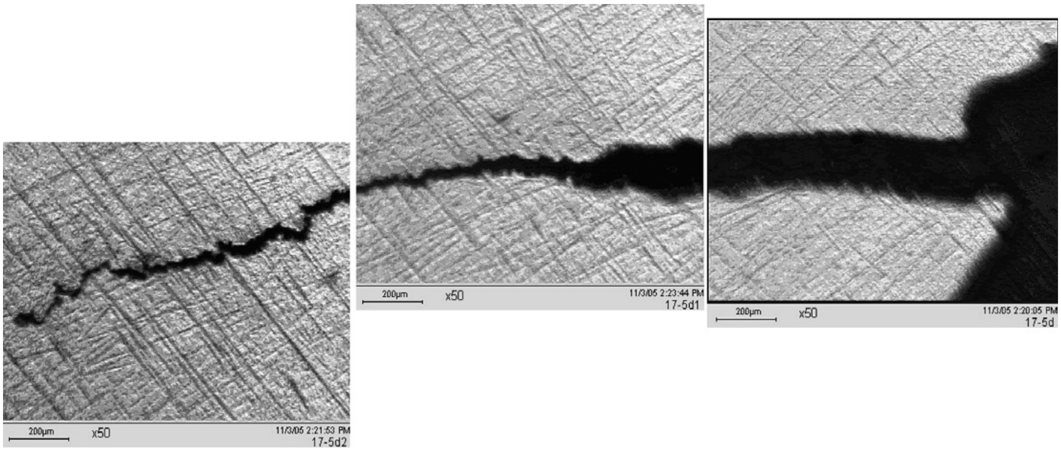

Fig. 3. Micrographes of an irradiated sample crack. Conditions: sample T15.

Table 2
Zr–2.5%Nb irradiated samples Ramberg–Osgood coefficients Young's modulus E and coefficients experimental fitting constants α and n

Sample	Temperature (K)	E (MPa)	α (–)	n (–)
15T	343	**101 269**	**0.3**	6.12
1TM	343	**101 269**	**0.22**	14.0
12L	423	**98 925**	**0.24**	17.4
16T	423	**98 925**	**0.24**	13.4
2TM	423	**98 925**	**0.23**	16.2
6TM	423	**98 925**	**0.24**	19.0
13L	523	**94 555**	**0.26**	14.5
17T	523	**94 555**	**0.25**	17.3
3TM	523	**94 555**	**0.22**	24.1
7TM	523	**94 555**	**0.22**	19.0
14L	573	**91 770**	**0.27**	12.7
18T	573	**91 770**	**0.26**	15.6
4TM	573	**91 770**	**0.23**	23.3
8TM	**573**	91 770	0.23	81.6

Table 2 values. All the coefficients show a characteristic temperature dependence, with the exception of specimen 8 TM.

4.4. Finite element analysis of the pressure tube for fracture mechanics parameters assessment

A stress–strain analysis for a Zr–2.5%Nb pressure tube was performed using the FEA-Crack computer code [19]. Material properties resulting from the data processing of the irradiated samples were used.

Semi-elliptic crack model dimensions used were: a crack depth (radial direction) c of 1 mm and a crack length (longitudinal direction) $2b$ of 10 mm.

The elasticity modulus as a function of temperature was taken from Table 2 for the irradiated samples 15–18T, which are considered as characteristic.

The four mechanical test temperatures (343, 423, 523 and 573 K) define the pressure tube operation range and the neutron fluence of 3.5×10^{20} cm^{-2} ($E > 1$ MeV), reached by the samples is that where saturation occurs for the mechanical properties of the irradiated Zr-2.5%Nb pressure tube.

For modelling reasons the dimensional data for the pressure tube were: an inside diameter R_I of 50.5 mm, a wall thickness t of 4 mm and an inside pressure P_{max} of 15 MPa (in the structural integrity analysis the inside nominal pressure is 1.5 P_{nom}).

In the situation when the hydrogen equivalent concentration is less than 35 ppm, and the neutron fluence Φ is in the range 1.8×10^{20} to 9.8×10^{21} cm^{-2} ($E > 1$ MeV), fracture toughness of the Zr–2.5%Nb alloy has the following temperature dependence [19]

Table 3
Mechanical characteristics and Ramberg–Osgood coefficients of the Zr–2.5%Nb irradiated samples for FEA-Crack code

Sample	Temperature (K)	$\sigma_{0\ real}$ (MPa)	$\sigma_{0.2real}$ (MPa)	UTS$_{real}$ (MPa)	E (MPa)	α (–)	N (–)
15T	343	**653**	**901**	**1138**	**101 269**	**0.3**	6.12
16T	423	**808**	**867**	**983**	**98 925**	**0.24**	13.4
17T	523	**760**	**835**	**871**	**94 555**	**0.25**	17.3
18T	**573**	700	803	820	91 770	0.26	15.6

$$K_{IC} = 32.3 + 0.022(T - 273) \quad (13)$$

with T in K and K_{IC} in MPa m$^{1/2}$.

The finite element analyses were performed with the FEA-Crack computer code for two property models: deformation plasticity and incremental plasticity. For this reason the Ramberg–Osgood parameters was selected as shown in the Table 3.

The data processing of the results obtained with the FEA-Crack code is taken into account as well as the aspect of von Mises mechanical stress field that suggests yield in the crack neighbourhood. The code predicts K and J parameters for structural integrity.

4.5. K_I and J_I mechanical parameters

As previously mentioned, internal coolant pressure for FEA-Crack analysis is 15 MPa to predict structural integrity in the 343–573 K range with an inside crack in the radial longitudinal plane.

Table 4
von Mises maximum stresses

Irradiated sample	Temperature (K)	$\sigma_{\text{von Mises}}$ (deformation) (MPa)	$\sigma_{\text{von Mises}}$ (incremental) (MPa)
15T	343	**643**	680
16T	423	**672**	674
17T	523	**654**	658
18T	**573**	637	642

Table 5
Stress intensity factor K_I maximum values

Sample	Temperature (K)	K_I (max) (deformation plasticity) (N mm$^{-3/2}$)	K_I (max) (incremental plasticity) (N mm$^{-3/2}$)
15T	343	**407.47**	407.34
16T	423	**407.23**	407.23
17T	523	**407.35**	407.35
18T	**573**	407.48	407.48

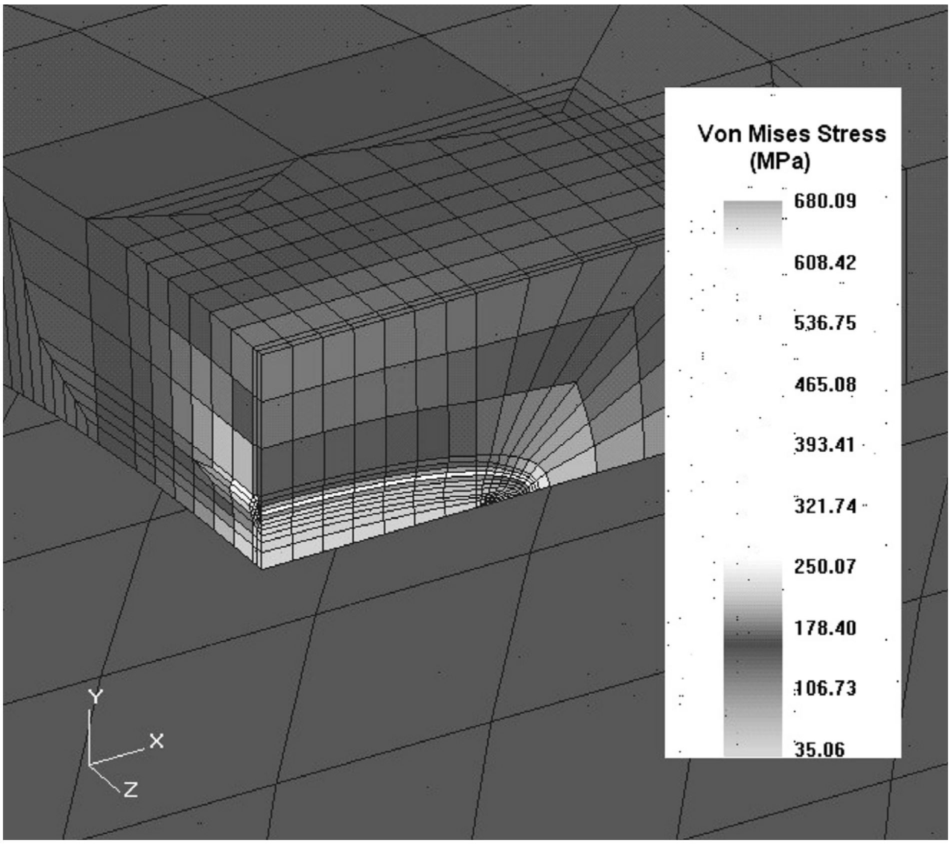

Fig. 4. von Mises stresses field for a semi-elliptic crack with a crack depth (radial direction) c of 1 mm and a crack length (longitudinal direction) $2b$ of 10 mm in 15T sample at 343 K modeled in FEA-Crack for incremental plasticity.

Table 6
Toughness parameter K_r and limit load parameter L_r coordinates of FAD for irradiated pressure tube with semi-elliptic crack ($P_{int} = 15$ MPa)

Irradiated sample	Temperature (K)	K_{IC} (MPa m$^{1/2}$)	K_I (MPa m$^{1/2}$)	K_r (–)	$L_{r\ global}$ (–)	$L_{r\ local}$ (–)
15T	343	**27.8**	**12.87**	**0.462**	**0.48**	0.50
16T	423	**29.6**	**12.88**	**0.435**	**0.50**	0.52
17T	523	**31.8**	**12.88**	**0.410**	**0.54**	0.56
18T	**573**	32.9	12.88	0.390	0.54	0.56

In Fig. 4 the von Mises stress fields are presented for crack depth (radial direction) c of 1×10^{-3} m and a crack length (longitudinal direction) $2b$ of 1×10^{-2} m at 343 K. The maximum stress values for deformation plasticity in the crack area are smaller compared with incremental plasticity for all the above mentioned temperatures.

In Table 4 the von Mises maximum stress deformation plasticity and incremental plasticity obtained by FEA-Crack for both material properties models are presented.

FEA-Crack post processes data and extracts mechanical fracture parameters: the intensity factors of K_I stresses from the J integral. Only the K_I maximum values were selected as representative, as shown in Table 5.

The K_I factors are quite similar, given the two modes of the material properties. They are the same both for 343 and 573 K. These values can be explained by the mechanical stresses in the crack adjacent area being in the elastic range and yield being limited to the crack tip. For this reason, K_r in the structural integrity analysis will use the K_I factor resulting from the incremental plasticity model which is more realistic for the material behaviour.

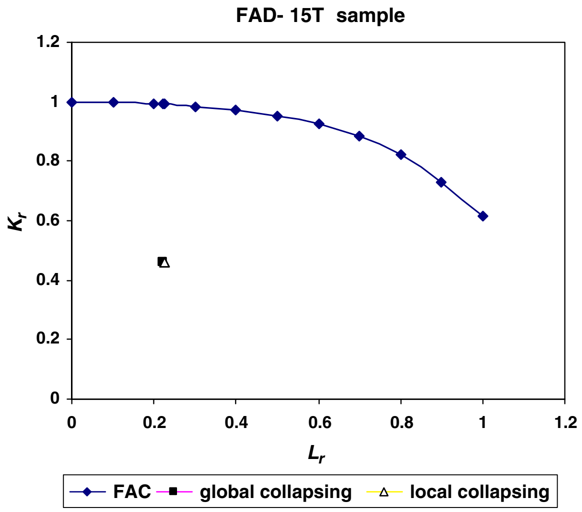

Fig. 5. Assessment points for semi-elliptic crack ($a = 1$ mm, $2c = 10$ mm) in the FAD diagram (option 1-R6/rev.5) for T15 probe at 343 K with global collapsing and local collapsing models.

K_r and L_r are presented in Table 6 for the analyzed crack for both types of collapsing pressure prediction. In Fig. 5, assessment points for specimen 15T are presented. For each situation, assessment points fall within the FAC safety area. This situation represents the case of pressure tube 'safe operation' at the above mentioned temperature.

5. Conclusions

The paper has presented the results of the fifth irradiation campaign with the irradiation capsule.

The irradiation of Zr–2.5%Nb pressure tube samples, with neutron fluence up to 3.5×10^{20} cm^{-2} ($E > 1$ MeV) was successfully achieved prior to performing tensile tests in the PIEL to obtain mechanical characteristics. These results were used for assessing the structural integrity of the pressure tubes used for Cernavoda Nuclear Power Plant (NPP).

The pressure tube structural integrity assessment methodology for flaws was presented.

A sharp flaw type, like a crack, in the radial-longitudinal plane of the CANDU pressure tube inside surface was modelled using the capabilities of the FEA-Crack computer code.

FEA-Crack computer code models for materials properties were made using the characteristics obtained from transversal direction irradiated samples 15–18T in the temperature range of 343–573 K. von Mises stresses were determined successfully. They describe irradiated alloy yield around the cracks for an inside pressure of 15 MPa and the increment plasticity model was found to have higher maximum von Mises stresses compared with deformation plasticity model.

It was found that maximum von Mises stresses decrease with temperature.

K_I and J mechanical fracture parameters of a pressure tube cracked on the inside surface were obtained as a function of mechanical load along the crack front for the test temperature range.

The stress intensity factor K_I had quite similar values both for incremental yield and deformation yield. For the temperature range 343–573 K, K_I coefficient did not alter its values significantly.

Structural integrity assessment according to [11] was made using both global collapsing and local collapsing models. The toughness parameter K_r values, which assess the crack initiation, were found to decrease with temperature increase, due to the fact that irradiated material toughness increase with temperature increase.

The local collapsing L_r values, which give the collapsing trend, increase with the increase of the temperature that can be explained by the material softening. The local collapsing L_r values were found to be greater than global collapsing L_r values.

The assessment points fall within the failure assessment curve (FAC) safety area and subsequently the analyzed cases are 'safe operation' situations. Structural integrity assessment methodology is a useful tool in assessing the behaviour of the CANDU reactor pressure tubes surveyed by the in-service inspections.

Acknowledgement

This work was done on behalf of Romanian Nuclear Authority (CNCAN) and Cernavoda Nuclear Power Station. The presentation of this work at EMRS 2008 was supported by the IAEA cooperation program.

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