

Hydrogen influence on fracture mechanics characteristics of zirconium alloy Zr-2.5 Nb at ambient and elevated temperatures

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1. Introduction

The analysis of fuel channels (FC) tube material aging influence on its service lifetime is executed from the positions of fuel channel ability to perform the predicted by design functions. It is well known, that metallic materials, especially titanium and zirconium alloys, accumulate hydrogen, which ingress effects mechanical and fracture characteristics of the materials.

Radiation strengthening and hydrogen embrittlement of the zirconium alloy. Hydrogen absorption also leads to the material embrittlement and delayed hydride cracking (DHC) and is one of the factors, which have determinant influence on tube service lifetime of the fuel channel. Therewith it is well known that increase of the metal temperature causes decreasing of main mechanical and fracture toughness characteristics.

Hydrogen influence on fracture toughness and J integral of Zr-2.5Nb zirconium alloy was investigated both at room and at elevated temperatures. There are some problems which should be resolved in order to provide fracture testing on hydrogen containing specimens. The first problem is related to the manufacturing of the specimens from the irradiated material. Due to tube irradiation it is impossible to test the specimens cut of directly from the used channel tube. The second problem is the preparation of hydrogen containing specimens. Manufacturing of the specimens in laboratory conditions requires special equipment and this procedure is a long time process. And the third problem is related to the testing technique, because curved specimens cut of from fuel tube have cylindrically shaped form and testing of such specimens is not clear yet from the positions of testing procedure, which is presented and described in the fracture mechanics testing standards.

In cooperation with Lithuanian Energy Institute curved compact specimens (ASTM E 399) were produced and containing at various hydrogen concentration levels (from 52 up to 140 ppm) in order to examine the influence of temperature and hydrogen concentration on fracture toughness characteristics K_{Ic} , K_{Ic}^* and critical J -integral value J_{Ic} . Hydrogen free and hydrogen containing specimens were tested in the temperature range 20 - 300°C and fracture criteria J_{Ic} has been determined by using load versus crack displacement and J - R curves testing methods.

2. Specimens and testing technique

Fracture toughness and J integral tests have been carried out using seminatural compact specimens cut of directly from fuel channel tube with the diameter of 88 mm and wall thickness of 4 mm. Therefore, parallel side sur-

faces of the specimens, alike tube wall, have cylindrically shaped form.

Shape and dimensions of the specimen is shown in Fig. 1. Mechanical characteristics of Zr-2.5Nb (TMT-2) alloy which corresponds with fracture test conditions are presented in the Table 1 [1].

The investigation of temperature and hydrogen concentration level influence on fracture toughness characteristics and J integral has been carried out on hydrogen free specimens (it is assumed that in the initial state after production FC the content of hydrogen in the zirconium pipe comprises 0.7 – 3 ppm) and on the specimens containing three hydrogen concentration levels: 52, 100 and 140 ppm.

Table 1
Mechanical characteristics of Zr-2.5Nb zirconium alloy

T, °C	Mechanical characteristics					
	$R_{p0.2}$, MPa	R_m , MPa	σ_f , MPa	A_5 , %	Z, %	$E \times 10^4$ MPa
Hydrogen free specimens						
20	411	492	608	14.78	62.2	3.44
150	288	374	514	13.95	70.7	2.90
300	230	293	533	11.06	74.4	3.16
Hydrogen concentration 52 ppm						
20	368	480	763	14.5	55.5	3.97
200	257	352	473	14.8	73.3	3.15
Hydrogen concentration 100 ppm						
20	470	545	967	13.08	62.9	4.72
170	280	322	727	13.52	75.4	4.88
300	252	305	494	12.62	78.8	3.19
Hydrogen concentration 140 ppm						
20	470	555	919	12.83	58.4	4.41
170	284	355	777	12.92	74.8	3.44
300	232	294	595	12.50	79.5	3.42

Testing has been performed on the 50 kN capacity experimental tension-compression testing machine with the stress σ_1 rate 20 MPa/s, which is in accordance with the requirements [2] to keep stress loading rate in the limits 2 – 20 MPa/s. The time of loading - unloading cycle at J integral investigation was approximately 40 s and the displacement rate was 1 mm/min.

In order to maintain normal tensile stress as perpendicular as possible to the crack growth plane during fracture toughness testing, the grips of compact specimens were subjected by the conical shape pins, which angle was calculated with respect to arch radius of the specimen and initial crack length, which has been precracked on high frequency testing equipment.

estimated by substituting F_Q by F_C in equation 2 (see Fig. 3).

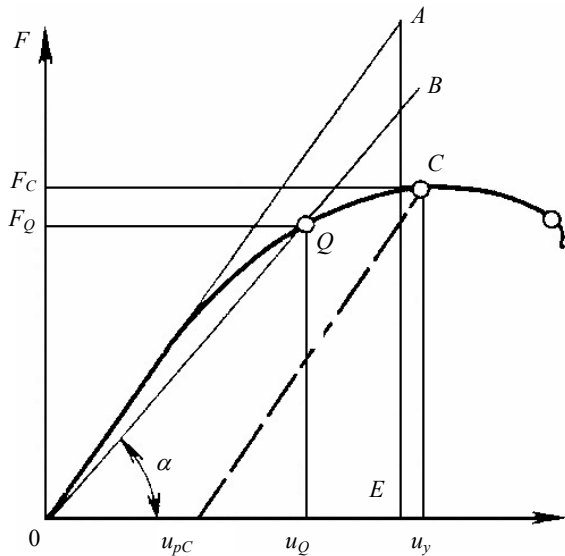


Fig. 3 Load versus crack displacement of the fourth type curves: a - scheme of F_Q and F_C determination

Table 2

Fracture toughness characteristics of Zr-2.5Nb alloy

$T, ^\circ\text{C}$	F_Q, N	F_C, N	$K_Q, \text{MPa m}^{1/2}$	$K_C^*, \text{MPa m}^{1/2}$
Hydrogen free specimens				
20	1954	3582	34.29	62.08
200	1065	2501	18.71	43.95
300	1705	2655	29.82	46.43
Hydrogen concentration 52 ppm				
20	1802	2893	31.15	50.09
200	892	2654	15.79	46.98
Hydrogen concentration 100 ppm				
20	1725	2880	29.79	50.03
170	1900	2897	33.73	51.12
300	860	2070	14.92	35.91
Hydrogen concentration 140 ppm				
20	1812	2610	31.76	45.64
170	1347	2553	23.77	45.07
300	1093	2057	19.10	35.92

The estimated stress intensity coefficients are shown in Table 2 and on Fig. 4. Stress intensity coefficients K_Q and K_C^* depends more upon testing temperature than upon hydrogen content. For example K_Q at the increase of temperature from 20 to 200°C for the specimens without hydrogen decreased from 34.29 to 18.71 $\text{MPa m}^{1/2}$, at saturation with hydrogen to 52 ppm – from 31.15 to 15.79 $\text{MPa m}^{1/2}$ and at saturation with hydrogen to 140 ppm – from 31.76 to 23.77 $\text{MPa m}^{1/2}$. At the increase of temperature up to 300°C for the specimen containing with hydrogen to 140 ppm K_Q decreased to 19.10 $\text{MPa m}^{1/2}$. Coefficient K_Q little depended on saturation quantity of hydrogen at 20°C.

Analogically K_Q at the increase of temperature altered K_C^* , at increase of hydrogen content from 0 to

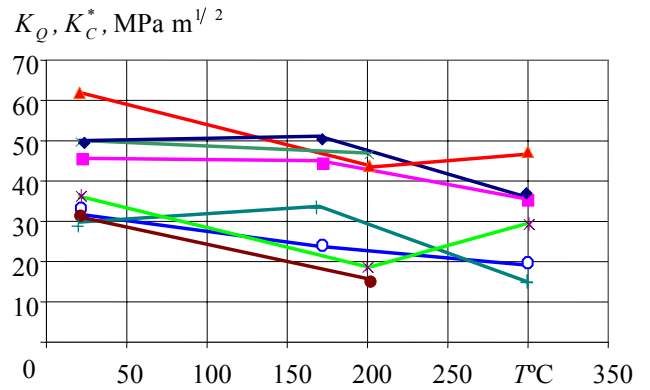


Fig. 4 Dependence of fracture toughness characteristics on saturation of hydrogen and temperature. K_Q * - 0 ppm, • - 52 ppm, + - 100 ppm, ○ - 140 ppm. K_C^* :▲ - 0 ppm, × - 52 ppm, ◆ - 100 ppm, ■ - 140 ppm

140 ppm at 20°C decreased K_C^* from 62.08 to 45.64 $\text{MPa m}^{1/2}$, however in temperature interval 170-300°C depended little on the quantity of hydrogen.

4. Experimental investigation of J integral

Using the test method described in [4-6], diagrams load versus crack opening displacement ($F - \delta$) by unloading–reloading cycles were recorded in order to define compliance of the specimens. Experimental curves “force – crack opening displacement” were recorded to computer via oscilloscope; load, displacement and compliance values at all loading-unloading segments were calculated by using scales $m_F = 10010 \text{ N/V}$ and $m_e = 2.123 \text{ mm/V}$.

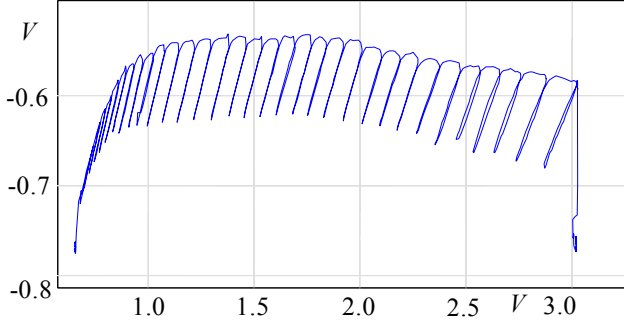
According to the testing program were tested 8 hydrogen free specimens (3 at 20°C, 2 at 170°C, 3 at 300°C), containing with hydrogen to 52 ppm 1 specimen at 20°C, containing with hydrogen to 100 ppm 3 specimens (in ones at 20, 170, 300°C), containing with hydrogen to 140 ppm (2 at 20°C, 3 at 170°C and 2 at 300°C). In total 19 specimens were tested.

During analysis of the results, given J integral values J_Q , J_{\max} and $J_{P_{\max}}$ for the same loading temperature and hydrogen concentration were averaged.

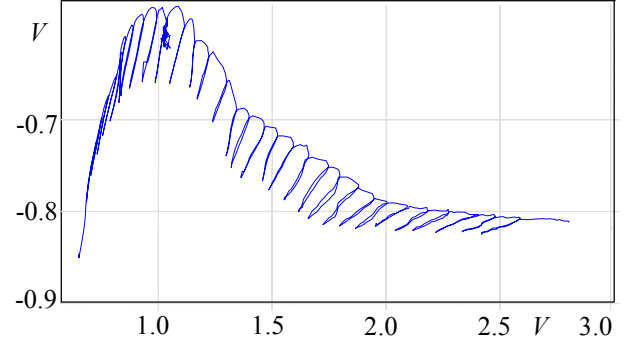
As follows from the Fig. 5, 6, more than 30 unloading–reloading cycles (standard requires minimum 10 cycles) for these specimens were executed. It should be noticed that compliance calculation is very sensitive process, because the angles of the adjacent segments, from which the compliance is calculated tilt is very negligible values.

Comparison of $F - \delta$ curves for hydrogen free specimens at 20, 170 and 300°C temperatures is presented in Fig. 5 and for hydrogen containing specimens to 100 ppm hydrogen concentration, at the same testing temperatures – in Fig. 6.

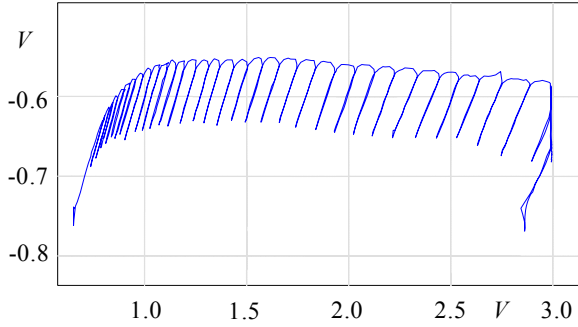
In order to evaluate temperature and hydrogen concentration influence on J integral critical values J_{IC} , at all $F - \delta$ the record points were calculated according to standard technique [5, 7].



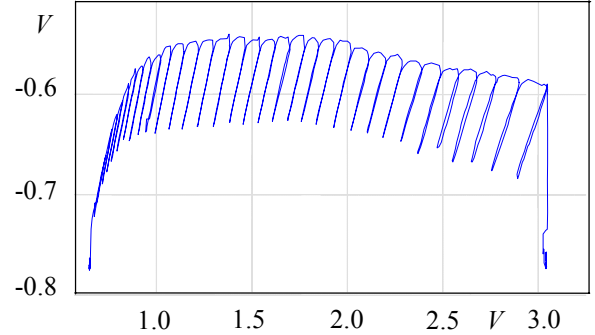
a



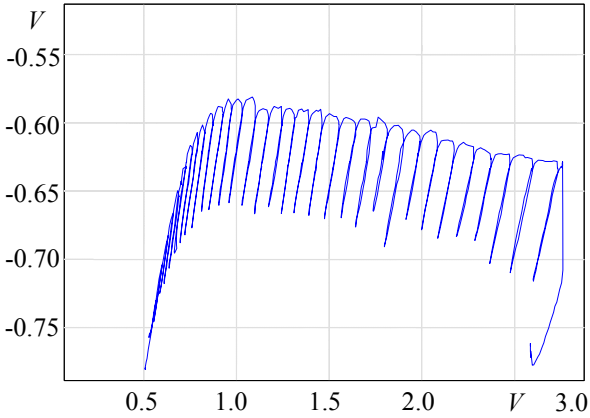
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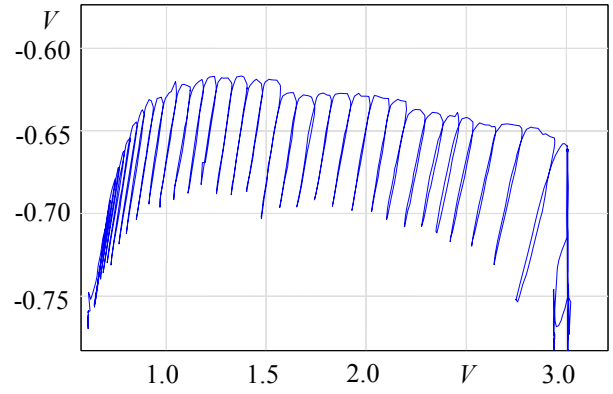
b



b



c



c

Fig. 5 Examples of “force – crack opening displacement” curves record for hydrogen free specimens: a - $T=20^{\circ}\text{C}$; b - $T=170^{\circ}\text{C}$; c - $T=300^{\circ}\text{C}$

Fig. 6 Examples of “force – crack opening displacement” curves record for hydrogen containing to 100 ppm specimens: a - $T=20^{\circ}\text{C}$; b - $T=170^{\circ}\text{C}$; c - $T=300^{\circ}\text{C}$

According to [4,5] J - R curves were established by smoothly fitting the data points to a power law regression line expressed as follows:

$$J = c_1 (Aa_p)^{c_2} \quad (3)$$

where c_1 and c_2 are parameters of the equation of region, limited by the given J_{max} and Δa_{max} and the exclusion line

$$J = 2\sigma_Y \Delta a_p \quad (4)$$

led out from the point $\Delta a_p = 0.15$ mm.

The maximum crack extension capacity was calculated by the equation

$$\Delta a_{max} = 0.1b_0 \quad (5)$$

The J integral value J_Q was defined at the intersection between the J - R curve (Eq. 3) and the exclusion line (Eq. 4) lead out from point $\Delta a_p = 0.20$ mm. If J_Q values satisfy the condition

$$b_0 \geq \frac{25J_Q}{\sigma_Y} = B^* \quad (6)$$

it is assumed that $J_Q = J_{IC}$.

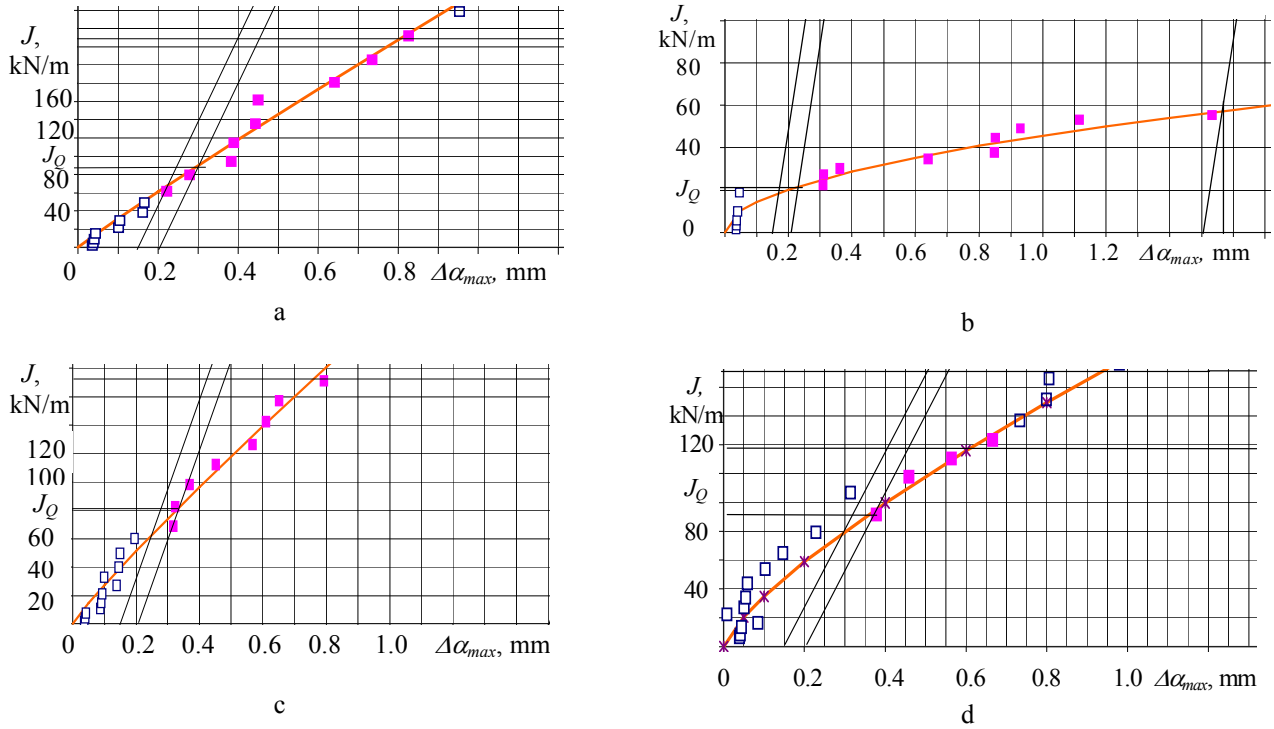


Fig. 7 Calculation of J_Q from J - R curve: a - $H_2=0$, $T=20^\circ\text{C}$; b - $H_2=140$ ppm, $T=20^\circ\text{C}$; c - $H_2=140$ ppm, $T=170^\circ\text{C}$; d - $H_2=140$ ppm, $T=300^\circ\text{C}$

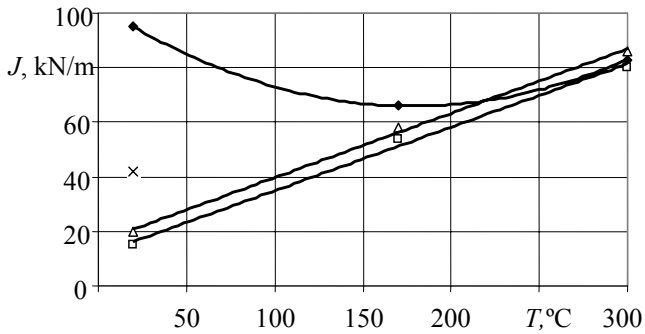


Fig. 8 Temperature influence on J -integral value J_{IC} :
 ◆ - $H_2=0$; × - $H_2=52$ ppm; Δ - $H_2=100$ ppm;
 □ - $H_2=140$ ppm

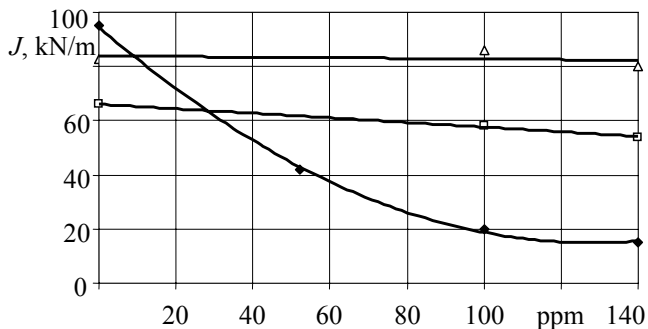


Fig. 9 Hydrogen concentration influence on J -integral value J_{IC} : ◆ - $T=20^\circ\text{C}$; □ - $T=170^\circ\text{C}$; Δ - $T=300^\circ\text{C}$

Examples of J - R curve construction [4, 5] and calculation procedure of J_Q on hydrogen free and containing specimens at normal and elevated temperatures are shown in Fig. 7.

Results of J integral calculation are presented in Table 3.

Table 3
Results of Zr-2.5Nb alloy J integral calculation

$T, ^\circ\text{C}$	$J_Q, \text{ kN/m}$	$J_{max}, \text{ kN/m}$	$J_{Pmax}, \text{ kN/m}$	$\Delta\alpha_{max}, \text{ mm}$
Hydrogen free specimens				
20	95	237	162	2.57
170	66	173	182	2.4
300	83	134	78	2.2
Hydrogen concentration 52 ppm				
20	42	219	76	1.78
Hydrogen concentration 100 ppm				
20	20	267	84	1.63
170	58	156	111	2.97
300	86	279	84	2.26
Hydrogen concentration 140 ppm				
20	15	266	31	1.56
170	54	169	88	2.14
300	104	135	121.5	2.58

Analysis has shown that the highest J_{IC} values were found for hydrogen free specimens at ambient (20°C) temperature and comprises 95 kN/m. Meanwhile the influence of temperature on hydrogen containing specimens gave opposite results: for the specimens containing up to 100 ppm concentration maximal J_{IC} was given at 300°C and comprises $J_{IC} = 86$ kN/m; at 170°C J_{IC} was given 58 kN/m; minimal J_{IC} value was given at 20°C and comprises 20 kN/m. The same character of J_{IC} variation is noticed for the specimens containing up to 140 ppm, however in this case J_{IC} values were lower in comparison to the results given on 100 ppm concentration specimens and comprises correspondingly: at 20°C $J_{IC} = 15$ kN/m; at 170°C - $J_{IC} = 54$ kN/m and at 300°C - $J_{IC} = 104$ kN/m (Figs. 8, 9).

4. Analysis of fracture mechanics characteristics

Satisfactory correlation of low cycle loading characteristics and mechanical ones, i.e. modified plasticity $Z_{mod} = (R_{p0.2}/R_m)Z$ [7-9] were determined in our earlier works.

In order to examine which mechanical characteristic of the alloy in the best way correlates fracture mechanics characteristics, various mathematical models were analyzed. It was found that modified plasticity criterion, expressed in the form Z_{mod} better describes K_C^* and J_{IC} in comparison with mechanical characteristics $R_{p0.2}, R_m, \sigma_f, Z$. Correlation of K_C^* and J_{IC} to Z_{mod} ($R=-0.61$ for K_C^* and $R=0.69$ for J_{IC}) and 95% confidence interval range (dotted lines) to the theoretical line is presented in the Figs. 10,11.

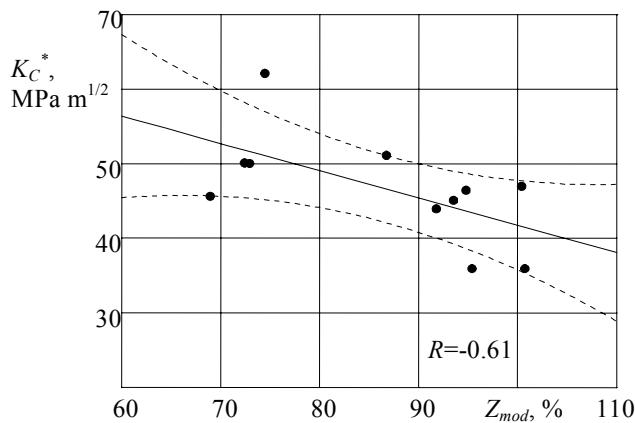


Fig. 10 K_C^* correlation to modified plasticity criterion Z_{mod} and its probabilistic evaluation

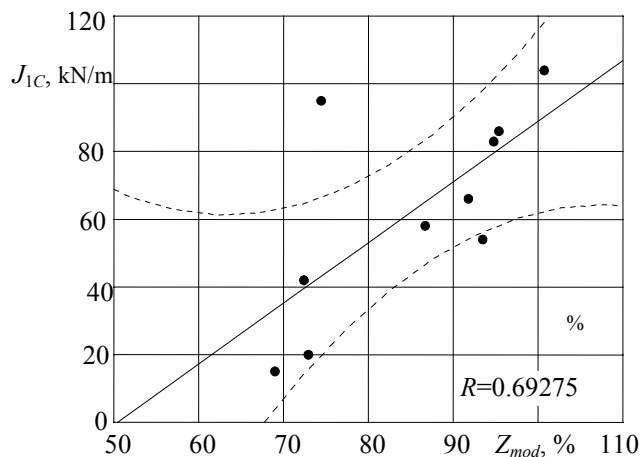


Fig. 11 J_{IC} correlation to modified plasticity criterion Z_{mod} and its probabilistic evaluation

Therefore, Z_{mod} could be taken as fracture criteria of Zr-2.5Nb zirconium alloy, which approximates satisfactory, the influence of temperature and hydrogen concentration on K_C^* and J_{IC} variation.

5. Conclusions

The investigation of temperature and hydrogen

concentration influence on fracture mechanics characteristics of fuel channel tube material Zr-2.5Nb zirconium alloy has provided the following results.

1. For fracture toughness the temperature factor is more significant than hydrogen presence. This statement was assessed for all tests, except the test on hydrogen containing specimens at 200°C, where the influence of hydrogen on K_C^* at the same temperature was given different by in comparison to the hydrogen free specimens. It means that the resistance to crack growth hydrogen containing zirconium alloy at 200°C is better than at room temperature.

2. The comparison of J_{IC} for hydrogen free and hydrogen containing specimens has shown that hydrogen concentration level, especially 100 and 140 ppm significantly reduces J_{IC} : for hydrogen free specimens at 20°C was $J_{IC} = 95$ kN/m, whereas for hydrogen containing specimens to 100 and 140 ppm J_{IC} correspondingly was 20 kN/m and 15 kN/m.

3. Fracture testing of the specimens, containing to 100 ppm at elevated temperatures has shown the increase of J_{IC} in comparison to the testing at 20°C from 20 kN/m to 58 kN/m at 170°C and to 86 kN/m at 300°C. Much more increase of J_{IC} is given for the specimens containing to 140 ppm, where at 20°C J_{IC} comprises 15 kN/m, at 170°C - 54 kN/m and at 300°C - 104 kN/m.

4. Under the temperature influence J_{IC} for the specimens containing to 100 and 140 ppm increases correspondingly to temperature elevation. For example - for hydrogen containing specimens up to 140 ppm at 20°C J_{IC} comprises 15 kN/m, at 300°C - 104 kN/m.

5. Analysis of K_C^* and J_{IC} dependence on mechanical characteristics of zirconium alloy has shown that modified plasticity Z_{mod} approximates satisfactory the influence of temperature and hydrogen concentration on K_C^* and J_{IC} variation, therefore Z_{mod} could be accepted as Zr-2.5Nb zirconium alloy fracture criterion.

References

1. **Daunys M., Dundulis R., Krasauskas P.** Hydrogen influence on zirconium alloy Zr-0.5Nb mechanical characteristics at ambient and elevated temperatures. -Mechanika. -Kaunas: Technologija, 2007, Nr.3(65), p.12-16.
2. European standard 10002-1. Metallic materials. Tensile testing – Part 1. Method of test at ambient temperatures, 2001.
3. GOST 25.506-85. Mechanical test methods of metals. Determination of fracture toughness characteristics under static loading, - Moscow, 1985 (in Russian).
4. Standard Test Method for Determining J - R Curves. ASTM E1152-87.
5. Standard Test Method for J_{IC} . A Measure of Fracture Toughness, Philadelphia, ASTM E813-87.
6. **Anderson, T.L.** Fracture Mechanics. Fundamentals and Applications. -Boca Raton: -CRC Press, 1991. -793p.
7. **Daunys, M.** Strength and Fatigue Life under Low Cycle Non-Stationary Loading. -Vilnius: Mokslas, 1989. - 256p. (in Russian).

8. Šniuolis, R., Daunys, M. Methods for determination of low cycle loading curves parameters for structural materials. -Mechanika. -Kaunas: Technologija, 2001, Nr.3(29), p.11-16.
9. Daunys, M., Šniuolis, R. Statistical evaluation of low cycle loading curves parameters for structural materials by mechanical characteristics.-Nuclear Engineering and Design, 2006, v.236, p.1352-1361.

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VANDENILIO ĮTAKA CIRKONIO LYDINIO Zr-2.5 Nb IRIMO CHARAKTERISTIKOMS APLINKOS IR PAAUKŠTINTOSE TEMPERATŪROSE

Darbo tikslas – ištirti Ignalinos AE antrojo energetinio bloko kuro kanalų vamzdžių, pagamintų iš cirkonio lydinio senėjimo procesą, t.y. nustatyti vandenilio koncentracijos ir temperatūros įtaką šio lydinio mechaninėms, irimo sąsūmos charakteristikoms ir kritiniam J_{1C} integralui.

Vienas iš pagrindinių faktorių sukeliančių eksploatacijos metu kuro kanalų medžiagos degradaciją yra vandenilis, kuris korozijos proceso metu yra absorbuojamas cirkonio lydinio. Esant vandeniliui cirkonio lydinyje pradėję formuotis hidridai sukelia šio lydinio sutrapėjimą ir tuo pačiu sumažina jo atsparumą trapiam irimui.

Irimo sąsūmos charakteristikos K_Q , K_C^* ir J integralo kritinės reikšmės J_{1C} kompaktiniams bandiniams ($B \approx 4$ mm) be vandenilio ir esant vandeniliui (52, 100 ir 140 ppm) nustatyta 20, 170, 200 ir 300°C temperatūrose.

Irimo sąsūmos charakteristikos mažai priklauso nuo vandenilio kiekio ir daugiau nuo – temperatūros. Kritinis integralas J_{1C} bandiniams turintiems vandenilio sumažėja 20°C temperatūroje, tačiau didėjant bandymo temperatūrai iki 300°C taip pat didėja.

Analizuojant cirkonio lydinio K_C^* ir J_{1C} priklausomybę nuo mechaninių charakteristikų nustatyta, kad modifikuotas plastiškumas $Z_{mod} = (R_{p0,2}/R_m)Z$ patenkinamai aproksimuoja temperatūros ir vandenilio įtaką šioms charakteristikoms.

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HYDROGEN INFLUENCE ON ZIRCONIUM ALLOY Zr-2.5 Nb FRACTURE MECHANICS CHARACTERISTICS AT AMBIENT AND ELEVATED TEMPERATURES

S u m m a r y

The main objective of the work – investigation of aging process of Ignalina NPP fuel channel tubes manufactured from zirconium alloy Zr-2,5 Nb, i.e. determination of the influence of hydrogen content and temperature on fracture toughness characteristics and critical integral J_{1C} .

The main factor initiating degradation of the fuel channel materials under operating conditions is hydrogen, which forms during corrosion process and is absorbed in

zirconium alloy. In zirconium alloy containing with hydrogen form hydrides, which cause brittleness of this alloy and decrease strength to brittle fracture.

Fracture toughness characteristics K_Q , K_C^* and J_{1C} were investigated on compact specimens ($B \approx 4$ mm) without hydrogen and containing with hydrogen (52, 100 and 140 ppm) at 20, 170, 200 and 300°C.

The investigation showed that characteristics of fracture toughness depended a little on hydrogen saturation value and more on temperature. The critical integral J_{1C} for containing with hydrogen specimens decreases at 20°C, however at the increase of test temperature to 300°C also increases.

Analysis of K_C^* and J_{1C} dependence on mechanical characteristics of zirconium alloy has shown, that modified plasticity $Z_{mod} = (R_{p0,2}/R_m)Z$ satisfactory approximated the influence of temperature and hydrogen concentration on this characteristics variation.

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ВЛИЯНИЕ ВОДОРОДА НА ХАРАКТЕРИСТИКИ МЕХАНИКИ РАЗРУШЕНИЯ СПЛАВА ЦИРКОНИЯ Zr-2.5 Nb ПРИ НОРМАЛЬНОЙ И ПОВЫШЕННЫХ ТЕМПЕРАТУРАХ

Цель работы – исследование процесса старения труб топливных каналов второго энергетического блока Игналинской АЭ изготовленных из циркониевого сплава, т.е. определение влияния концентрации водорода и температуры на характеристики вязкости разрушения и критический интеграл J_{1C} .

Один из основных факторов вызывающих деградацию материала топливных каналов во время эксплуатации является водород, который абсорбирует сплав циркония. При наличии водорода в сплаве циркония образовавшихся гидриды, вызывают его окрупчивание и уменьшают сопротивление хрупкому разрушению.

Характеристики вязкости разрушения K_Q , K_C^* и J_{1C} исследовались на компактных образцах ($B \approx 4$ мм) без водорода и при наличии водорода до 52, 100 и 140 ppm при нормальной и повышенных до 170, 200 и 300°C температурах.

Проведенные исследования показали, что характеристики вязкости разрушения мало зависят от количества водорода и больше – от температуры, а J_{1C} для образцов имеющих водород уменьшается при 20°C, но при повышении температуры до 300°C возрастает.

Анализ зависимости K_C^* и J_{1C} от механических характеристик показал, что модифицированная пластичность $Z_{mod} = (R_{p0,2}/R_m)Z$ удовлетворительно описывает влияние температуры и водорода на характеристики разрушения.

Received March 5, 2007

DOI: 10.5755/j02.mech.14838