Hydrogen influence on zirconium alloy Zr-0.5Nb mechanical characteristics at ambient and elevated temperatures

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1. Introduction

The analysis of material aging influence on service lifetime of fuel channels (FC) tube is executed from the positions of fuel channel ability to perform predicted by the design its functions. It is well known that radiation in certain degree can have an effect on all functions and properties of fuel channel material. Radiation may cause changes in fuel channel tube sizes; in addition to this, an essential effect on the majority of functions and properties of the fuel channel tube has radiation strengthening, embrittlement and hydrogen ingress.

Hydrogen ingress leads to the embrittlement and degree hydride cracking (DHC) of zirconium alloy and is one of the factors, which has an influence on fuel channel tube service lifetime. Also it is one of the consequences of corrosion, which depends on the parameters of heat-transfer agent and structural state of material.

Usually the analysis of kinetic dependence of the hydrogen ingress in the zirconium alloy is examined in connection with the kinetics of oxidation, but to analyze the kinetics of oxide film is very difficult because it is washed away by coolant.

During reactor operation, several factors influence FC aging including hydrogen ingress to zirconium material, embrittlement, corrosion and the related reduction of pipe wall thickness, possible crack growth by DHC mechanism when hydrides are present in the alloy.

Aging mechanisms and fuel channel service lifetime influencing factors are presented in Table 1.

Table 1

Aging mechanisms and fuel channel service lifetime influencing factors

Aging mechanisms	Influencing factors on FC lifetime
Radiation strengthening and embrittlement	Change of mechanical properties
Hydrogen absorption	Hydrogen embrittlement and delayed hydride cracking (DHC)
Corrosion	Change of pipe wall thickness
Thermal and radiation creep under static loading	Change of pipe measurements

Thus, the main factors limiting fuel channel service lifetime are operational regimes such as temperature and irradiation, which action is influencing on the change of mechanical characteristics of tube material.

Taking this in to account, the investigation of hydrogen concentration level and temperature influence on zirconium alloy Zr-2.5Nb (TMT-2) mechanical characteristics was carried out.

Hydrogen free and hydrogen saturated tensile specimens with hydrogen concentration levels 52, 100 and 140 ppm were tested at normal (20ºC) and elevated (150, 170, 200 and 300ºC) temperatures.

The carried out tests on monotonous tension of the specimens enabled to calculate the main mechanical characteristics of zirconium Zr-2.5Nb alloy: stress of proportional limit σ_{pl}, yield strength $R_{p0.2}$, ultimate strength R_m, fracture stress σ_f, strain of proportional limit e_{pl}, relative elongation A_5, area reduction Z and modulus of elasticity E.

2. Specimens and testing technique

As the pipe strength depends mainly on circumferential stresses, seminatural specimens in this direction from the fuel channel 88 mm diameter and 4 mm wall thickness tube, were cut off. Shape and dimensions of the specimens are shown in Fig. 1.

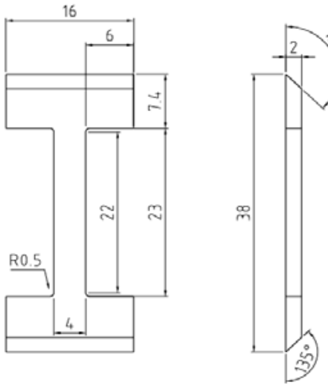


Fig. 1 Shape and dimensions of tensile specimen

The investigation was performed on the 50 kN capacity tension-compression testing machine with the stress rate σ_1 =20 MPa/s, which is in accordance with the requirements [1] - to keep stress loading rate in limits σ_1=2-20 MPa/s. Experimental curves "force – specimen elongation" were recorded to computer via oscilloscope; load and strain values were calculated by using scales m_F =10010 N/V and m_e =2.667 mm/V.

Testing of the specimens at elevated temperatures was performed by using induction heating with predicted temperature control accuracy ±2.5ºC. In order to maintain initial hydrogen concentration level in the hydrogen saturated specimens, temperature increasing rate was set as possible slow (2-5ºC/min). An example of hydrogen saturated specimen heating record is shown in Fig. 2.

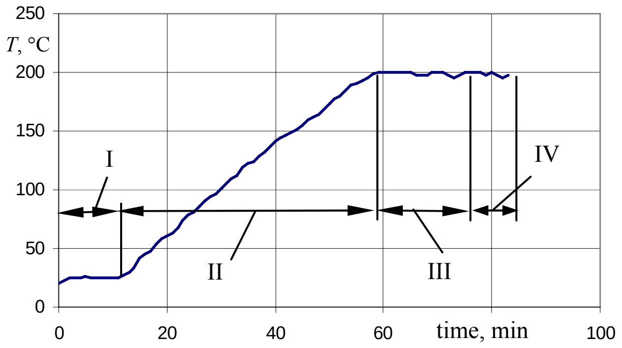


Fig. 2 Hydrogen saturated specimen heating record: I – starting stage; II – monotonic increase stage; III – stabilization stage; IV – testing stage

Tensile testing and calculation technique was performed according to EN-10002-1 and EN-10002-5 requirements [1, 2]. In detail, testing procedure is described in [3], however due to the specimen's shape nonconformity to the standard requirements, testing base of the specimens was calculated from the Eq. (1)

$$L_b = L_c + \left[0.005\, b\, r - \left(0.025\, b \right)^2 \right]^{0,5} \qquad (1)$$

where L_b is the length of specimen's rectangular cross-section , b is thickness of the specimen, r is testing base and fixing head radius (r=0.5 mm).

Relative elongation of the specimen after break was calculated according to the expression:

$$A_5 = \frac{L_{bf} - L_b}{L_b} \qquad (2)$$

where L_{bf} is the length of specimen's rectangular cross-section after it's break.

Calculation scheme of the specimen cross-section after its break is shown in Fig. 3. Reduction of area after break was calculated according the equation:

$$Z = \frac{a\, b - a_f\, b_f}{a\, b} \qquad (3)$$

where a and b is initial width and thickness of the specimen's testing zone cross-section, a_f and b_f is the width and thickness of the specimen after break correspondingly.

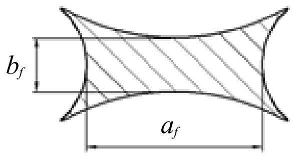
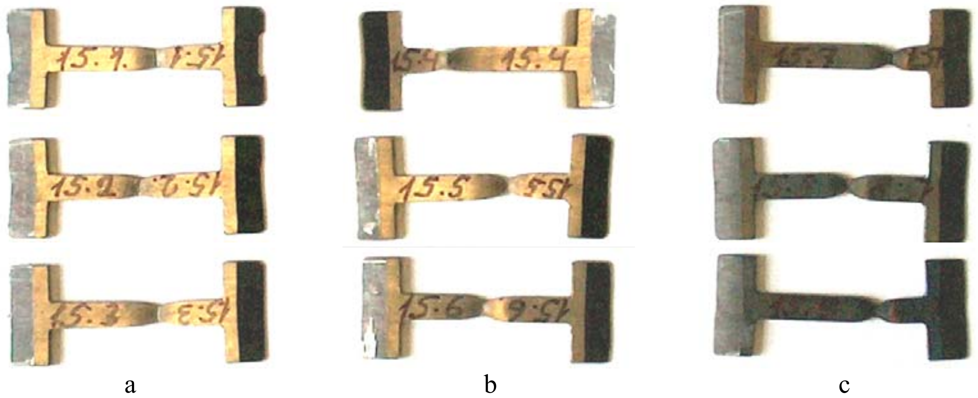


Fig. 3 Calculation scheme of the specimen cross-section reduction

Examples of the specimens after break saturated to 100 ppm hydrogen concentration at 20, 170 and 300ºC are shown in Fig. 4



a b c

Fig. 4 Photos of the specimens saturated to 100 ppm hydrogen concentration after fracture: a- 20ºC; b – 170ºC; c - 300ºC

3. Investigation of mechanical characteristics

According to the testing program 12 hydrogen free zirconium alloy specimens were tested: at 20ºC - 3 specimens, at 150ºC – 4 specimens and at 300ºC – 5 ones.

Hydrogen saturated to 52 ppm concentration level alloy specimens were tested at 20 and 200ºC only, so this test set comprises 8 (2×4) specimens, on hydrogen saturated to 100 and 140 ppm concentration levels at 20, 170 and 300ºC 18 (2×3×3) specimens were tested. Thus, during tensile testing, in total, 38 specimens were tested.

Temperature influence on mechanical characteristics of hydrogen free zirconium Zr-2.5Nb alloy specimens on monotonous tension is shown in Fig. 5 (here one curve is mean of all specimens' test in the set). The comparison of tension diagrams for the specimens saturated to 52, 100 and 140 ppm at elevated temperatures is presented in Fig. 6-8.

It is obviously seen from the graphs, that temperature increase from 20 to 300ºC led to the monotonous decrease of strength characteristics σ_{pl}, $R_{p0.2}$, R_m.

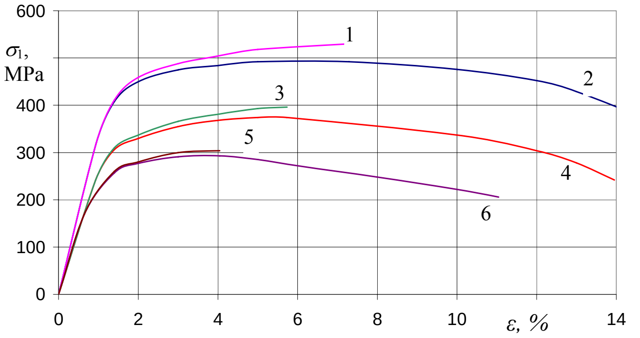


Fig. 5 Comparison of tension diagrams for hydrogen free specimens at 20ºC (1, 2), 150ºC (3, 4) and 300ºC (5, 6)

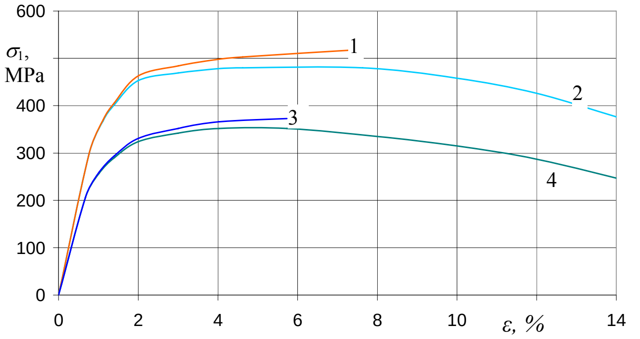


Fig.6 Comparison of tension diagrams for the specimens, saturated to 52 ppm hydrogen concentration at 20ºC (1, 2) and 200ºC (3, 4)

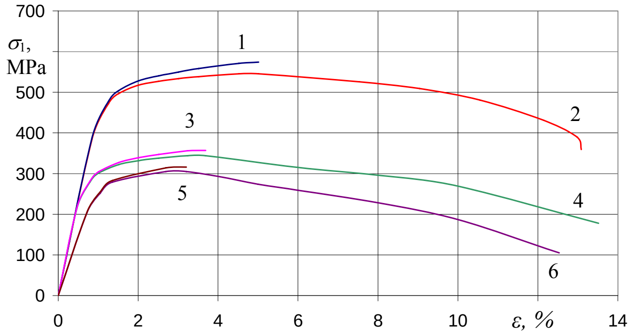


Fig. 7 Comparison of tension diagrams for the specimens, saturated to 100 ppm, at 20ºC (1, 2), 170ºC (3, 4) and 300ºC (5, 6)

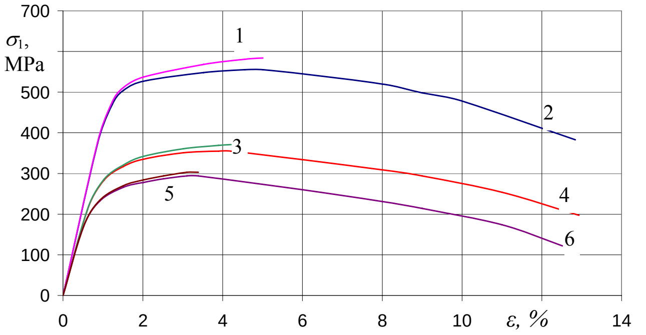


Fig. 8 Comparison of tension diagrams for the specimens, saturated to 140 ppm, at 20ºC, (1, 2) 170ºC (3, 4) and 300ºC (5, 6)

True and engineering tension curves are presented in these figures for each test set. Engineering stress-strain curves are calculated dividing force by initial cross-section of the specimen, true ones are constructed till the point corresponding ultimate strength force, because during the test, cross-section of the specimen reduces and therefore the force is divided by the current cross-section.

Main mechanical characteristics of the investigated zirconium Zr-2.5Nb alloy are presented in Table 2.

Table 2
Mechanical characteristics of Zr-2.5Nb zirconium alloy

T, ºC	Mechanical characteristics					
	$R_{p0,2}$, MPa	R_m, MPa	σ_f, MPa	A_5, %	Z, %	$E\times10^4$ 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

4. Discussion

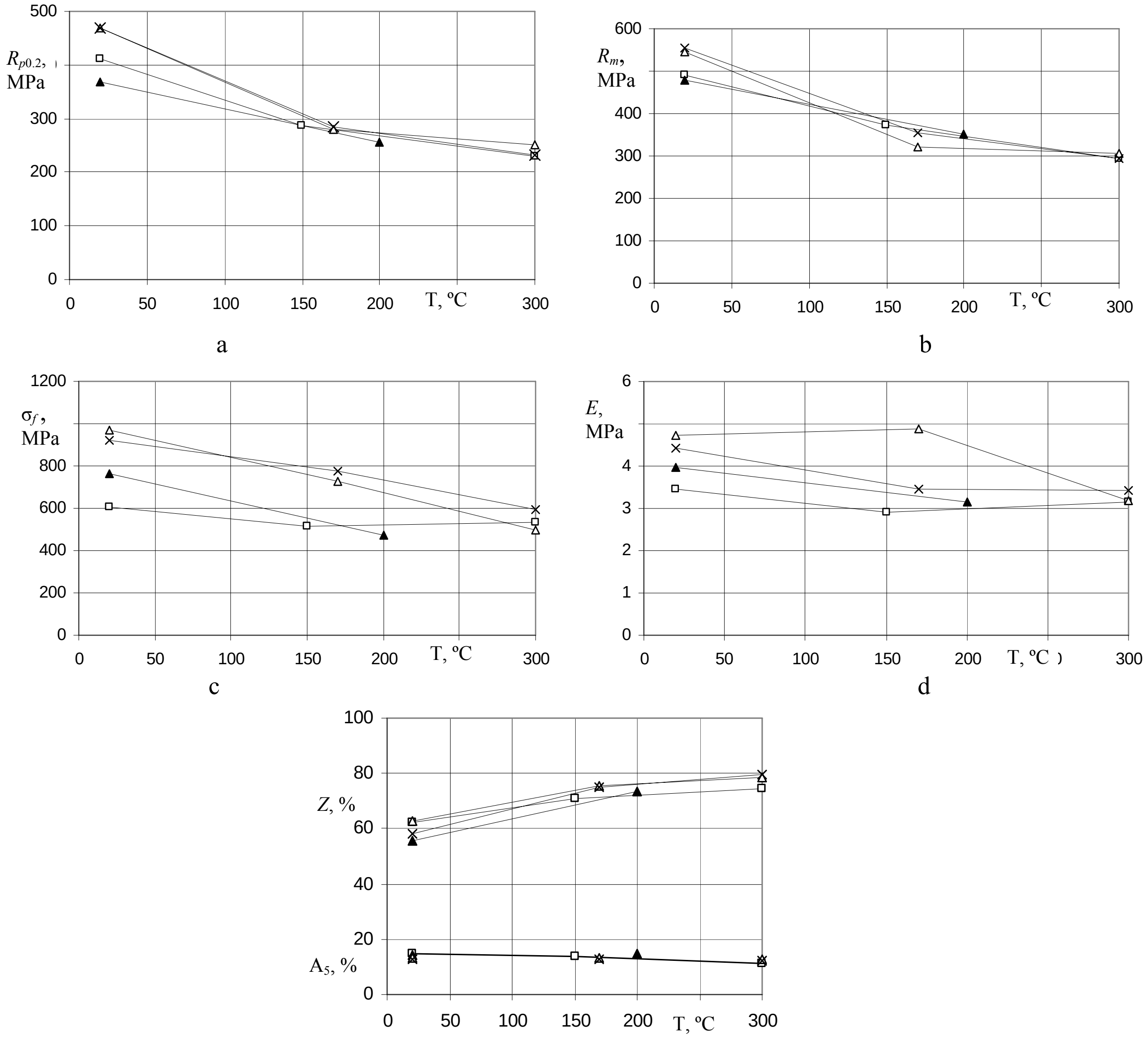
Experimental results on hydrogen concentration and temperature influence on zirconium alloy mechanical characteristics that are presented in the Table 2 are averaged values from the set of 3-5 specimens, repeated at the same testing conditions. The influence of temperature and hydrogen concentration on zirconium alloy strength mechanical characteristics $R_{p0,2}, R_m, \ \sigma_f$ experimental results were displayed graphically as shown in the Fig. 9, a, b, c.

Observation of the experimental data enabled to conclude that increasing of hydrogen concentration from 0 to 140 ppm at 20ºC temperature causes the increasing of strength characteristics: - $R_{p0.2}$ from 368 (52 ppm) to 470 MPa, R_m - from 480 (52 ppm) to 555 MPa and σ_f - from 608 to 919 MPa.

Hydrogen saturated specimen's tests at 150-200°C gave the following results: yield limit $R_{p0.2}$ for all hydrogen concentration levels remains constant, R_m marginally decrease from 374 (0 ppm) to 322 MPa (100 ppm), meanwhile fracture stress σ_f increase from 514 up to 777 MPa (140 ppm).

Observation of tests at 300ºC has shown that yield limit $R_{p0.2}$ and ultimate strength R_m remains stable for all hydrogen concentration levels.

Fracture stress of the alloy σ_f (Fig. 9, c) at all temperatures increases and together hydrogen concentration increase. So at 20ºC for hydrogen free alloy σ_f comprises 608 MPa, in comparison to σ_f =967 MPa for hydrogen concentration 100 ppm. The same tendency is no-



a

b

c

d

e

Fig. 9 Temperature influence on mechanical characteristics of zirconium Zr-2.5Nb alloy: a - yield strength $R_{p0,2}$; b- ultimate strength R_m ; c – fracture stress σ_f ; d - modulus of elasticity E ; e - relative elongation A_5 (upper graph) and reduction of the area *Z*. □ – hydrogen free specimens; ▲ – H_2=52 ppm; Δ - H_2=100 ppm; × - H_2=140 pp

ticed for the tests with other temperatures, but the difference in values is less, i.e. at 300ºC σ_f increases from 494 MPa (100 ppm) to 595 MPa (140 ppm).

Modulus of elasticity *E*, for hydrogen free specimens, as our experiment showed (Fig. 9, d) under temperature influence dereases from 34400 MPa at 20ºC to 31600 MPa at 300ºC. For specimens saturated to 52 ppm at ambient temperature modulus of elasticity is higher in comparison to the hydrogen free specimens however at 200ºC the difference is trivial. The most valuable increase of elasticity modulus is for the specimens saturated to 100 ppm at 170ºC temperature, however at 300ºC the modulus decreases again till hydrogen free alloy values level. For the specimens, saturated to 140 ppm temperature has the same influence like for 100 ppm test; however at 170 and 300ºC the modulus of elasticity is given slightly higher.

The analysis of zirconium alloy plasticity has shown that area reduction *Z* does not depend on hydrogen concentration and increases only under temperature influence (Fig. 9, e). For example, at 20ºC *Z* 62.2%, at 150ºC - 70.7%, meanwhile for hydrogen free specimens at 300°C the reduction of area increases up to 74.4%. The same character of plasticity alternation is noticed on the specimens with 52, 100 and 140 ppm hydrogen content. Meanwhile relative elongation A_5 practically does not depend on temperature and hydrogen concentration.

4. Conclusions

Investigation of temperature and hydrogen influence on mechanical characteristics of fuel channel tube material (Zr-2.5Nb zirconium alloy) has provided the following results.

1. Mechanical characteristics $R_{p0.2}, R_m, \sigma_f, E$ of the zirconium alloy were the highest at room temperature in comparison to 170, 200 and 300ºC, meanwhile plasticity characteristics - reduction of area *Z* is the highest at 300ºC whereas relative elongation A_5 remains stable.

2. Hydrogen concentration has lower influence on mechanical characteristics in comparison to the tem-

perature influence; only fracture stress σ_f for hydrogen saturated specimens at all investigated temperatures is considerably higher.

3. Modulus of elasticity E at 20-200ºC temperatures increases in relation to hydrogen concentration increase, but at 300ºC this effect disappears.

References

1. European standard 10002-1. Metalic materials. Tensile testing – Part 1. Method of test at ambient temperatures, 2001.
2. European standard 10002-5. Metalic materials. Tensile testing – Part 5. Method of test at elevated temperatures, 1991.
3. **Daunys, M., Dundulis, R., Krasauskas, P.** Investigation of RBMK-1500 reactor fuel channel material aging process.-Proc. of the Ninth International Conference on Material Issues in Design, Manufacturing and Operation of Nuclear Power Plants Equipment, Pushkin-St Petersburg, 6-8 June, 2006, v.1, p.155-163.

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VANDENILIO KONCENTRACIJOS ĮTAKA CIRKONIO LYDINIO Zr-2.5Nb MECHANINĖMS CHARAKTERISTIKOMS APLINKOS IR AUKŠTESNĖSE TEMPERATŪROSE

Reziumė

Straipsnyje pateikti vandenilio koncentracijos ir temperatūros įtakos Ignalinos AE 2-ojo energobloko RBMK-1500 reaktoriaus kuro kanalų medžiagos − cirkonio lydinio Zr-2.5Nb (TMT-2) mechaninėms charakteristikoms tyrimo rezultatai. Statinio tempimo bandymai atlikti naudojant plokščius stačiakampio skerspjūvio bandinius 20−300ºC temperatūrų intervale, atitinkančiame įvairius reaktoriaus darbo režimus. Bandiniai be vandenilio buvo bandyti 20, 150 ir 300ºC temperatūrose, o nuo 52 iki 140 ppm prisodrinti vandenilio − 20, 170 200 ir 300ºC temperatūrose. Tyrimas parodė, kad, didėjant temperatūrai, stiprumo mechaninės charakteristikos pratėja, o skerspjūvis siaurėja. Prisodrinimas vandenilio didesnę reikšmę šioms charakteristikoms turi tiktai 20-170ºC temperatūrų intervale ir didėja mažėjant temperatūrai.

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

Summary

This paper deals with the investigation of the influence of hydrogen concentration on zirconium alloy Zr-2.5Nb (TMT-2) mechanical characteristics at various operating regimes, which are related to temperature variation from normal to elevated up to 300ºC. Tensile specimens, which were cut off from the reactor fuel channel tube free and hydrogen saturated at concentration levels from 52 up to 140 ppm were tested in order to evaluate the influence of hydrogen ingress on mechanical properties of the alloy. Specimens made from hydrogen free alloy were tested at 20, 150 and 300ºC temperatures, while hydrogen saturated specimens were tested at 20, 170, 200 and 300ºC. Results of the investigation has shown that at temperature increase mechanical characteristics of strength decrease and reduction of are increase. Saturation with hydrogen has stronger influence on mechanical characteristics only at 20-170ºC and increases decreasing temperature.

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ВЛИЯНИЕ КОНЦЕНТРАЦИИ ВОДОРОДА НА МЕХАНИЧЕСКИЕ ХАРАКТЕРИСТИКИ СПЛАВА ЦИРКОНИЯ Zr-2.5Nb ПРИ КОМНАТНОЙ И ПОВЫШЕННЫХ ТЕМПЕРАТУРАХ

Резюме

В статье приведены результаты исследования влияния концентрации водорода и температуры на механические характеристики циркониевого сплава Zr-2.5Nb (ТМО-2), являющегося материалом топливного канала реактора RBMK-1500 2-го энергоблока Игналинской АЭС. Исследование механических характеристик проведено на плоских призматических образцах при статическом растяжении. Температура испытания была выбрана в интервале от 20 до 300ºC и соответствовала различным рабочим режимам реактора. Образцы без водорода испытались при температурах 20, 150 и 300ºC, а насыщенные различным уровнем концентрации водорода − от 52 до 140 ppm − при температурах 20, 170, 200 и 300ºC. Анализ результатов исследования показал, что с повышением температуры испытания прочностные механические характеристики сплава уменьшаются, а сужение поперечного сечения возрастает. Концентрация водорода существенное влияние на механические характеристики оказывает только в интервале температур 20-170ºC и увеличивается с понижением температуры.

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