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Effect of test temperature and load ratio on hydride cracking rate of Zr-2.5Nb alloy

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1. Introduction

Pressure tubes of Zr-Nb alloy are used in the reactor core to contain fuel assemblies. Over time, corrosion process leads to gradual pick up of hydrogen in the tubes. When hydrogen concentration in the tubes exceeds terminal solid solubility (TSS) in a certain temperature range [1], the tubes are susceptible to crack initiation and propagation. The process is called delayed hydride cracking (DHC). Cracking process is caused by hydrogen migration up to tensile stress gradient to the region of stress concentration. When the local TSS is exceeded, brittle hydride platelets precipitate normal to tensile stress. When hydride platelet reaches a critical length, it cannot support the concentrated stress and it ruptures. The crack advances this distance and the process repeats. This incremental growth of the crack forms striations on the fracture surfaces. There is an incubation period between each crack growth increment while a new hydride zone is formed at the crack tip [2]. It is known that there is a dependence between striation spacing, DHC velocity and yield strength.

The necessary conditions for DHC are the presence of a crack, a sufficiently high hydrogen concentration, tensile stresses and a stress intensity factor (K_I) larger than the threshold value at the crack tip. Threshold value condition is known as K_{IH}, - the critical stress intensity factor in the presence of hydride, below which no crack growth occurs. For most Zr-2.5Nb pressure tubes K_{IH} is in the range 5-10 MPa·m$^{1/2}$ [3]. At K_I values above K_{IH} the rate of cracking, V_{DHC}, is essentially independent of K_I.

Temperature dependence of DHC velocity in axial direction was determined for both CANDU and RBMK Zr-2.5%Nb pressure tubes. The influence of cyclic loading was investigated for RBMK material at 250°C. The need to evaluate the influence of variable loading on the DHC process is related with uncertainty about K_I values initiating DHC and the influence of cyclic loading on DHC growth rate in Zr-2.5Nb pressure tube materials.

2. Experimental procedures

Test specimens were prepared using both CANDU cold worked and RBMK (TMT-1 heat-treated condition) Zr-2.5Nb pressure tube materials.

Sections of the pressure tube were hydrided to a required hydrogen concentration using an electrolytic method and diffusion annealing treatment [4]. The hydrogen concentration in the samples were chosen so that solubility limit at the test temperature was exceeded about 25 ppm.

After adding hydrogen curved compact toughness (CCT) specimens with the width of 17 mm were machined along the axis of the pressure tube, retaining the tube curvature. The CCT specimens have been fatigue pre-cracked at room temperature to produce an initial crack length of about 1.7 mm

DHC testing procedures are based on IAEA recommendations [5].

Crack propagation process during DHC test has been monitored using potential drop (PD) versus crack length calibration curve. In all tests constant DC current of 8A was supplied to the specimens. Potential drop output was measured using the wire (diameter 0.5 mm) made from the same pressure tube material to eliminate stray EMF. The wires were attached to the specimen within 1 mm of either side of the notch by spot welding. PD versus time changes has been continuously recorded using computer data acquisition system. DHC test was completed after estimated crack growth reached 1.5 mm.

After completion of DHC test actual crack length was measured from fractographs. Fractured specimens were photographed using computerized image analysis system. On the digital image the DHC and fracture area boundaries were outlined manually by drawing one pixel width line, then the image was transformed to binary and corresponding areas have been measured.

The DHC velocity was determined from the average DHC-length measured from fracture surface photograph for one half of the fractured specimen. Crack length was estimated as a ratio of measured crack area and specimen width.

3. DHC test results

It is known [6], that maximum DHC value is attained by cooling from a temperature higher than the solvus temperature of hydrogen in the specimen. Because DHC velocity is sensitive to the temperature history, a standardized procedure for measuring the cracking velocity was applied. Specimens were heated at 5°C per min to a peak temperature T_{pk}, soaked for 1h and cooled down at 1.5°C per min to the test temperature T_{tst} with a minimum 30°C temperature drop without any under cooling. Peak and test temperatures for DHC velocity measurement are presented in Table 1. Temperature schedule for testing at 250°C is illustrated in Fig. 1. After holding the specimen at test temperature for 35 min, a constant static load sufficient to give an initial stress intensity factor of 15 MPa m^{½} was applied on the specimen, then after 15 min cycling loading device was launched. DHC started after a short incubation time and was allowed to grow to an estimated crack length. Test data, including crack length, incubation and cracking time, DHC velocity and loading parameters are given in the Table 2.

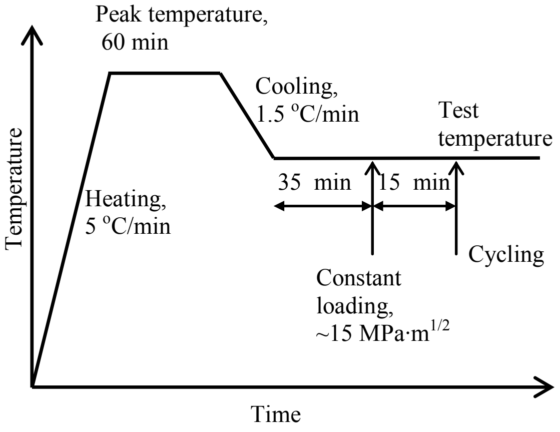


Fig. 1 Temperature-time schedule for DHC testing

Table 1

Peak and test temperatures for DHC testing

Pressure tube material	Peak temperature, °C	Test temperature, °C	Measured hydrogen concentration, ppm
CANDU	259	144	32
CANDU	315	250	62
CANDU	327	283	79
RBMK	275	144	27
RBMK	315	250	54
RBMK	327	283	76

Cyclic loading tests were done at 250°C with specimens containing 55 ppm of hydrogen. Two specimens from the same section were tested under constant loading conditions (R=1). Other specimens were tested under cycle loading at 1 cycle/min and a different load ratio R. Load ratio R was calculated as a ratio of minimum and maximum load in a cycle. The variation of loading during DHC test was recorded every 4 sec. to a computer data file. Minimum and maximum K_I values at start and at the end of test are presented in Table 2.

Temperature dependence of DHC velocity is shown in Fig 2. Figure 3 shows DHC axial velocity data at 250°C depending on load ratio R.

After completion of DHC test the specimen was unloaded and cooled down in the furnace to room temperature and fractured. Typical fractographs of specimens subjected to DHC test are shown in Fig. 4.

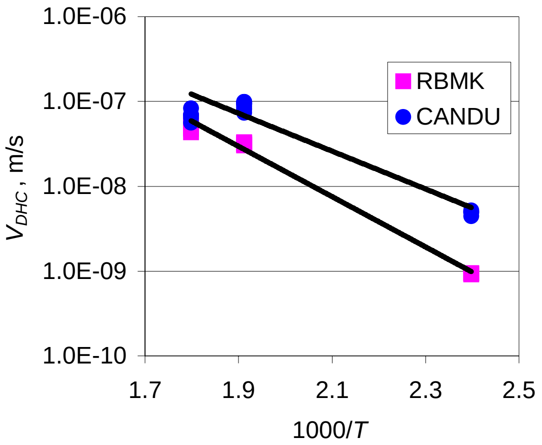


Fig. 2 DHC axial velocity data as a function of 1000/T

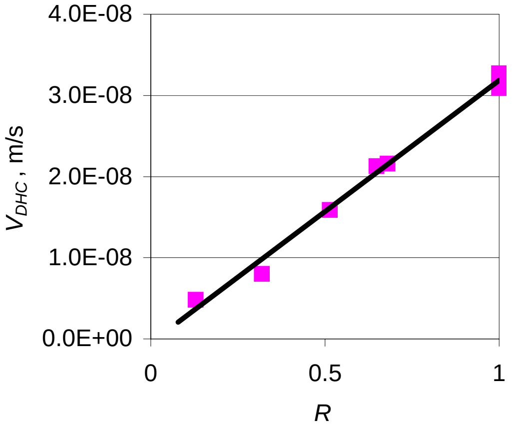
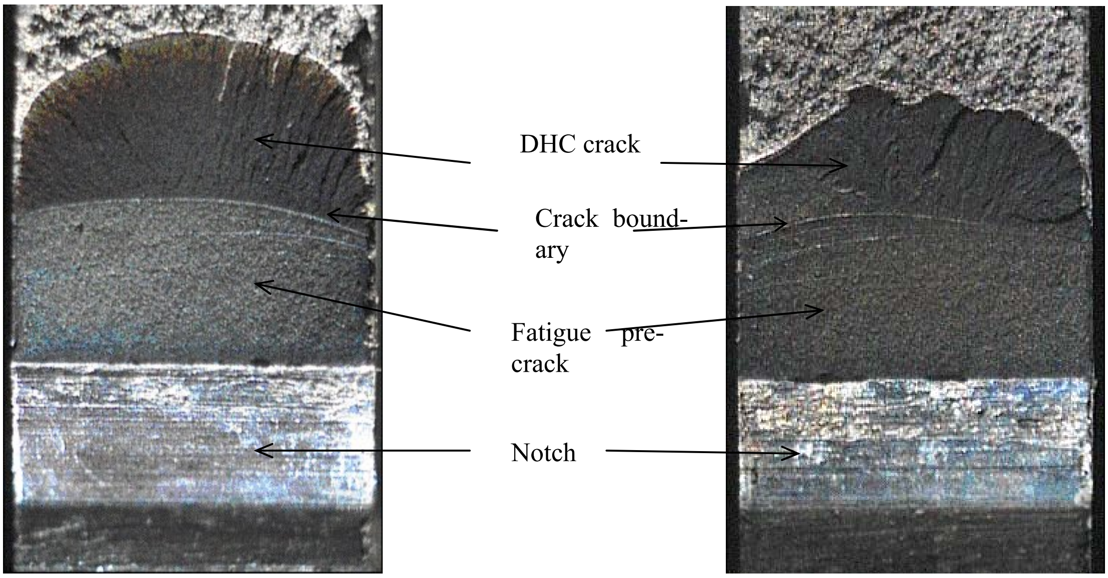


Fig. 3 DHC axial velocity as a function of load ratio R; test temperature 250°C

Table 2

Summary of test results for DHC velocity measurements at 250 °C (RBMK TMT-1)

Specimen I.D.	DHC crack length, mm	Incubation time, min	Cracking time, min	DHC velocity, m/s	Initial K_{Imin} / K_{Imax}, MPa·m$^{1/2}$	Final K_{Imin} / K_{Imax}, MPa·m$^{1/2}$	Load ratio R=P_{min}/ P_{max}
4.0	1.003	30	3473	4.81E-09	2.1/15.29	2.54/18.56	0.13
4.1	1.531	43	826	3.09E-08	15.68	21.25	1
4.2	1.123	253	527	3.27E-08	15.1	18.7	1
4.3	1.681	78	1220	2.16E-08	10.27/15.03	14.06/20.57	0.68
4.4	1.157	28	2411	8.00E-09	4.75/14.88	5.95/18.62	0.32
4.6	1.506	42	1583	1.59E-08	7.63/14.85	10.3/20.15	0.51
4.7	1.528	55	1195	2.13E-08	9.9/15.27	13.4/20.75	0.65



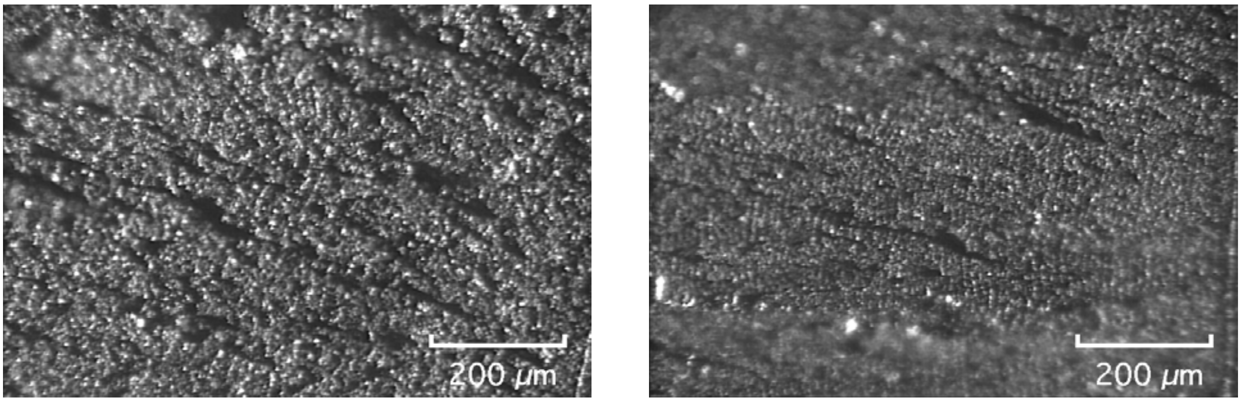
a b

Fig. 4 Photo of the fracture surface of CCT specimens, subjected to DHC testing at static (a) and cyclic (b) loading conditions at 250°C

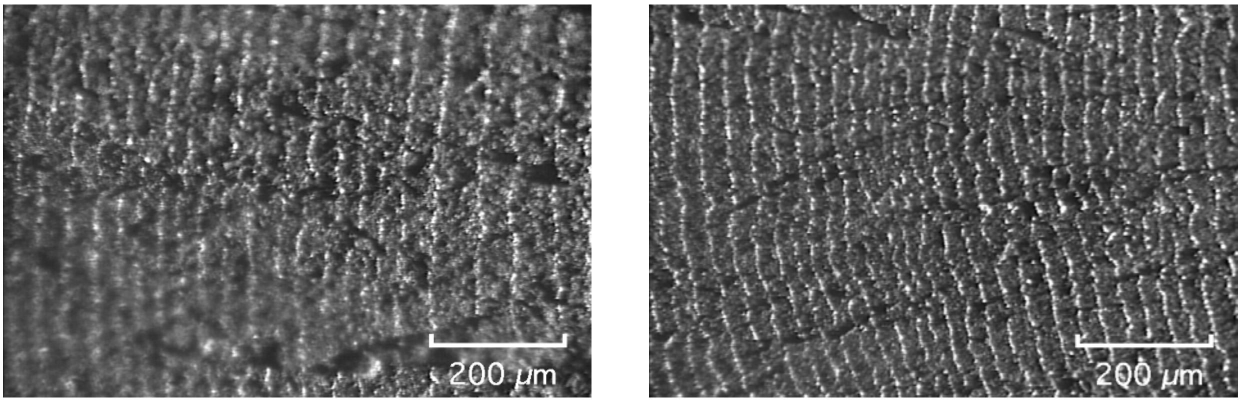
4. Discussion

The performed tests show that DHC rate depends on test material, temperature and loading conditions.

Testing under different load ratio R ($R=P_{min}/P_{max}$) shows that DHC rate decreases with R value 6.7 times, from $3.2\cdot10^{-8}$ m/s at constant loading to the lowest value, which is nearly $4.8\cdot10^{-9}$ m/s. During crack growth, ratio K_{Imin}/K_{Imax} remained constant, while maximum K_I values at the test end increased at the average 4.5 MPa $\mathrm{m}^{1/2}$. It can be observed a sharp increase in DHC rate (Fig. 7) when loading mode changes from static to cyclic, thereafter cracking rate gradually decreases. Subsequent crack growth is even and the slope of PD versus crack length curve remains constant during the test, as K_I values increases.

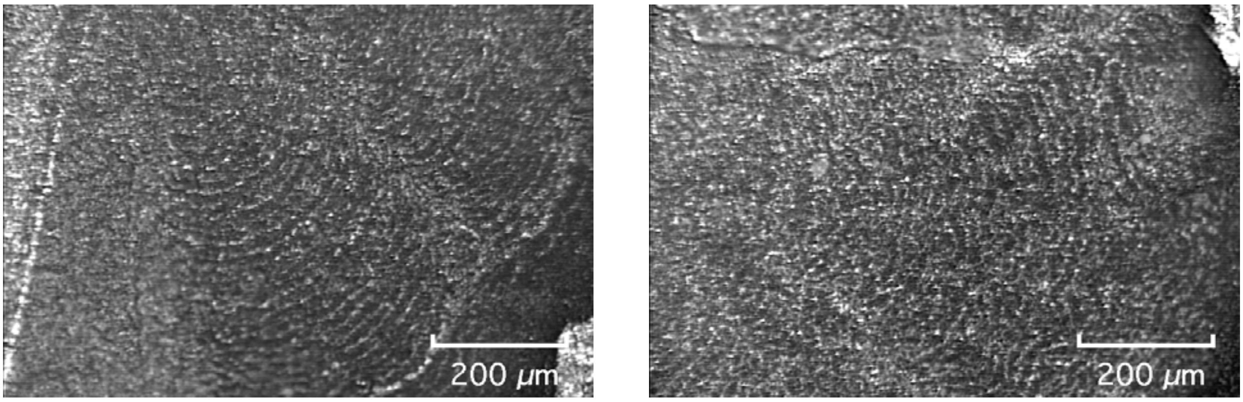


144°C, RBMK 144°C, CANDU

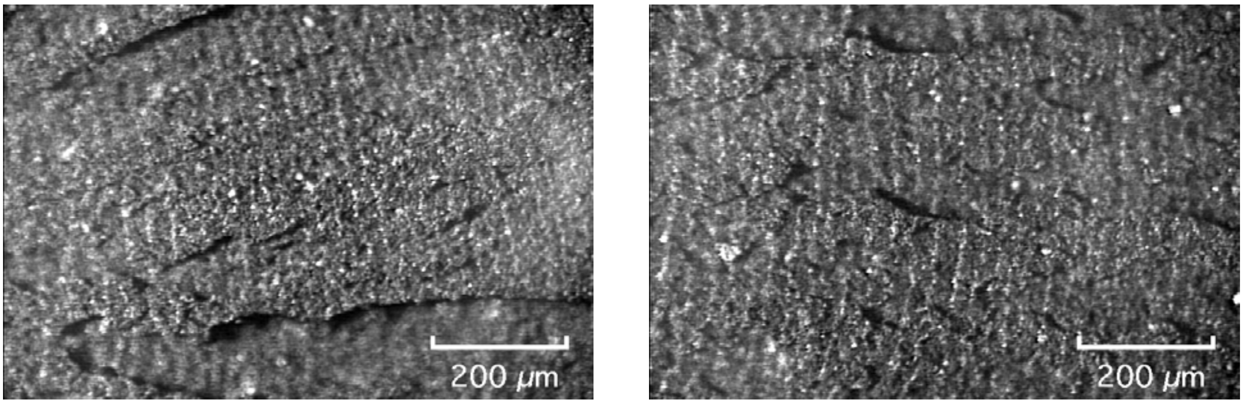


283°C, RBMK 283°C, CANDU

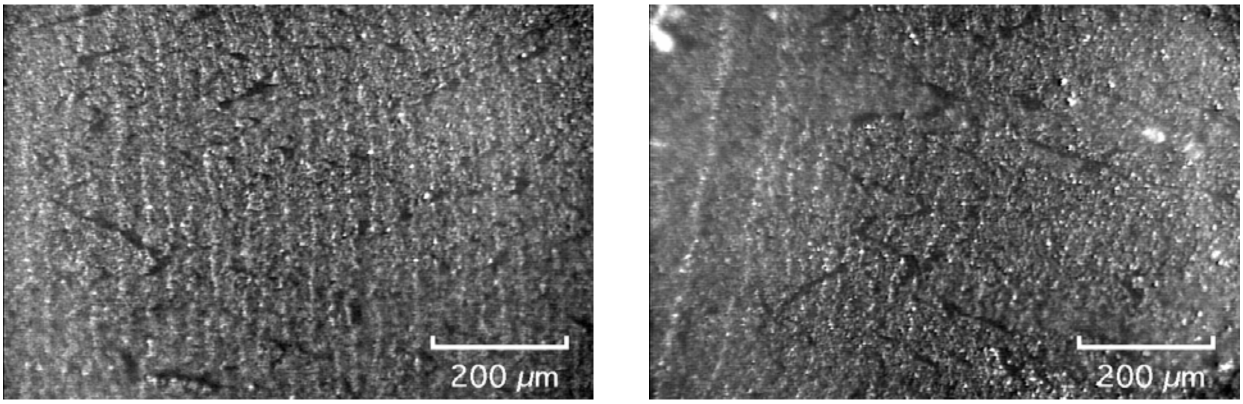
Fig. 5 Striations formed by DHC at different temperatures



R=0.129, Specimen 4.0 R=0.319, Specimen 4.4



R=0.514, Specimen 4.6 R=0.68, Specimen 4.3



R=1, Specimen 4.1 R=1, Specimen 4.2

Fig. 6 Photographs of DHC fracture surface after cyclic loading test. Test temperature 250°C

Consequently, DHC rate remains nearly constant and depends on R. DHC velocity, while R>0.1, and $K_{I\ max}$ <21 MPa $m^{1/2}$ follows relationship:

$$V_{DHC} = 3.24 \cdot 10^{-8} \cdot R - 5.15 \cdot 10^{-10}$$

Fabrication route influences microstructure, which as a result influences the strength of the alloy, terminal solid solubility and effective diffusivity of hydrogen. It is known [7] that RBMK pressure tube material has lower yield strength at 250°C compared to CANDU pressure tube. The differences that can affect DHC rate are grain structure, crystallographic texture and strength. Both microstructure and strength can independently affect crack velocity.

It was found that striation spacing increases with the increase in test temperature and are larger in the specimens of the RBMK material than in the CANDU material. Inter-striation spacing measured from the fracture photographs (Fig. 5, 6) are shown in Fig. 8. The likely explanation is associated with the decrease of matrix strength as temperature increases. DHC is an intermittent crack growth process, involving precipitation, growth and eventual of the hydride when it reaches critical length. Striation spacing is associated with the length of hydride [8].

Inter-striation spacing as well as DHC rate depends on the test temperature. In case of cycling loading DHC rate and inter-striation spacing decreases with load ratio R (Fig. 9). It can be observed that at low R-values, striations on DHC fracture surface almost disappear. Surface morphology also changes depending on R. When R=0.13 crack surface becomes smooth and a little differs from fatigue crack. Steps on the PD curve, which are characteristic for DHC growth under constant loading, disappear when R decreases (Fig. 10). There is a correlation between the number of striations on the fracture surface and the number of PD jumps [8]. The applied tensile stress

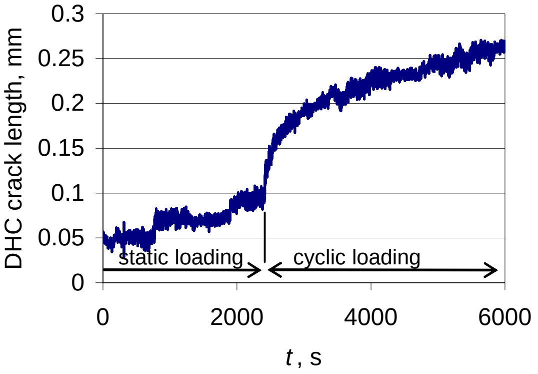


Fig. 7 Sharp increase of DHC rate at changing loading mode from static to cyclic

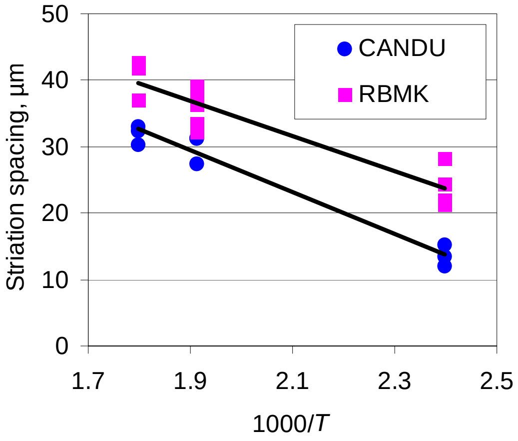


Fig. 8 Relationship between inter-striation spacing and temperature

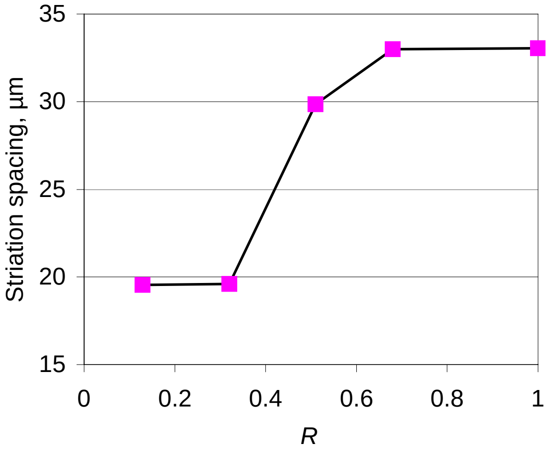


Fig. 9 Dependence of inter-striation spacing on the load ratio *R*; test temperature 250°C

to fracture hydrides is a function of hydride length [9].

It is known that the orientation of hydride platelets in zirconium alloys depends on texture [6]. The decrease of DHC rate under cyclic loading apparently can be related to the texture changes at the crack tip which determines different orientation of hydride platelets, as in the case of constant loading, when long hydrides are formed and their spatial orientation depends on the whole alloy texture. That also could explain differences in DHC surface morphology.

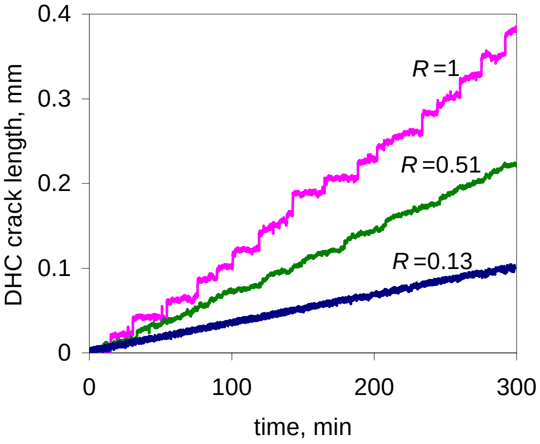


Fig. 10 Crack length versus time at different load ratio *R*

5. Conclusions

The measured DHC velocity depends both on temperature and on load ratio *R* and increases with the increase in test temperature. DHC velocities in RBMK samples, depending on temperature are 3-5 times lower than in CANDU pressure tube material. Decreasing load ratio *R* from 1 to 0.13, DHC rate for RBMK material at 250°C linearly decreases about 7 times from $3.2\cdot10^{-8}$ m/s to $4.8\cdot10^{-9}$ m/s.

Temporary, but considerable increase of DHC rate can be observed when loading mode changes from constant to cyclic. This process leads to a small increase of crack length up to 0.2-0.3 mm.

DHC rate as well as inter-striation spacing, measured from fracture surface photographs of the specimens, increases with test temperature when subjected to constant loading. In case of variable loading testing at constant temperature, DHC rate and inter-striation spacing decreases with load ratio.

Acknowledgments

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BANDYMO TEMPERATŪROS IR APKROVOS KOEFICIENTO ĮTAKA LYDINIO Zr-2.5Nb HIDRIDINIO PLEIŠĖJIMO GREIČIUI

Reziumė

Nustatytas lydinio Zr-2.5Nb lėto hidridinio pleišėjimo (LHP) greitis esant skirtingoms temperatūros ir apkrovos sąlygoms. Vandenilio turintys bandiniai buvo pagaminti iš kuro kanalų RBMK (TMO-1) ir CANDU vamzdžių, nusodinant hidrido sluoksnį bei homogenizuojant jį aukštoje temperatūroje iki reikiamos vandenilio koncentracijos.

Lėto hidridinio pleišėjimo greičiai esant pastoviai apkrovai išmatuoti 144, 250 ir 283°C temperatūrose. Nustatyta, kad LHP greitis, kylant eksperimento temperatūrai, didėja.

Bandymai, esant 1 ciklo/min apkrovos dažniui, atlikti 250°C temperatūroje. Nustatyta, kad mažėjant apkrovos asimetrijos koeficiento R reikšmėms nuo 1 iki 0.13, LHP greitis sumažėja iki 7 kartų - nuo $3.2\cdot10^{8}$ iki $4.8\cdot10^{9}$ m/s.

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EFFECT OF TEST TEMPERATURE AND LOAD RATIO ON HYDRIDE CRACKING RATE OF Zr-2.5Nb ALLOY

Summary

Delayed hydride cracking (DHC) rate in Zr-2.5Nb pressure tube material was determined under different temperature and loading conditions. Test specimens containing hydrogen were manufactured from RBMK (TMT-1) and CANDU pressure tube materials, depositing hydride layer and homogenizing it at elevated temperatures to the required hydrogen concentration.

Delayed hydride cracking rates at constant loading were measured at 144, 250 and 283°C.

Cycle loading tests were done at 250°C. Loading frequency was 1 cycle/min with different minimum/maximum load ratio R, changing from 0.13 to 1. It was determined that with the decrease of load ratio R, DHC rate increases up to 7 times, from $3.2\cdot10^{-8}$ m/s to $4.8\cdot10^{-9}$ m/s.

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ВЛИЯНИЕ ТЕМПЕРАТУРЫ ИСПЫТАНИЯ И КОЭФФИЦИЕНТА НАГРУЗКИ НА СКОРОСТЬ ЗАМЕДЛЕННОГО ГИДРИДНОГО РАСТРЕСКИВАНИЯ СПЛАВА Zr-2.5Nb

Резюме

Скорость замедленного гидридного растрескивания (ЗГР) в трубах топливных каналов РБМК (ТМО-1) и CANDU из сплава Zr-2.5Nb определялась при разных температурах и условиях нагрузки. Испытательные образцы наводорожены осаждением гидридного слоя с последующей его гомогенизацией при повышенной температуре до получения требуемой концентрации водорода.

Скорость замедленного гидридного растрескивания в условиях постоянной нагрузки измерена при температурах 144, 250 и 283°C. Установленно, что с увеличением температуры скорость ЗГР возрастает.

Испытания при циклической нагрузке 1 цикл/мин проведены при 250°C. Установлено, что с уменьшением величины коэффициента асимметрии нагрузки R от 1 до 0.13, скорость ЗГР уменьшается до 7 раз: от $3.2\cdot10^{-8}$ до $4.8\cdot10^{-9}$ м/с.

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