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Stress-induced Hydride Reorientation and Cracking in Fuel Cladding Tube

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The goal of this research was to find out if reorientation can influence cladding failure and determine conditions and possible mechanism of failure. Reorientation tests were done on zircaloy nuclear fuel cladding specimens. Specimens were made having two triangular notches along specimen axis, which are similar to the surface flaw that can form during operation. The loading was applied using pin-loading tension (PLT) loading fixture to give stress proportional to a distance from the specimen front. When loaded, specimen was cooled from 340 °C to 40 °C at a cooling rate 0.25 °C/min., total duration of test was 20 h. After testing, the specimen was examined layer by layer metallographically to determine hydride reorientation under certain stress conditions. In case of a surface flaw reorientation of hydrides takes place perpendicular to the stress direction. It was found that hydride reorientation occurs at a stress range (77 – 89) MPa, for initiation of delayed hydride cracking (DHC) process larger stress is required than for hydride reorientation.

Keywords: zirconium alloys, fuel cladding, hydride, reorientation.

1. INTRODUCTION

In some zircaloy nuclear fuel cladding, hydride cracking was found in form of long splits that allowed substantial leakage of fission products [1, 2]. Cladding in PWR, BWR, as well as in RBMK reactors is usually a tube made from zirconium alloy (Zircaloy-4 or Zr1Nb) with diameter of about 9 mm – 14 mm, wall thickness of about 0.6 mm – 0.9 mm and length of about 4 m. The design of the RBMK reactor fuel is similar to fuel elements manufactured for standard PWR or BWR-type reactors [3]. If the cladding wall is penetrated during operation, for example by fretting, water from the heat-transport system can enter into the fuel rod gap where steam is produced. Much hydrogen is generated because the steam oxidizes the fuel and the inside surface of the cladding, reducing the partial pressure of oxygen and leaving a gas rich in hydrogen. At some distance from the primary defect the gas becomes almost pure hydrogen, and with breakdown of the protective oxide layer, hydrogen may be absorbed by the cladding. With fuel expansion during power ramping, the hydrided cladding is stressed, which leads to crack initiation. The cracks grow through-wall and may be over 1 meter long [4]. The fractures are characterized by brittle regions in “striations” or “chevrons”, with the crack front often leading towards the outside surface of the cladding.

Integrity of the fuel claddings is also important during transportation and storage of spent fuel. The main factors, which can influence cladding integrity are creep, but hydride reorientation or the delayed hydride cracking (DHC) (Fig. 1), still exist with respect to the mechanical behaviour of spent fuel cladding in dry storage [5]. It is assumed [6], that mechanical properties and susceptibility to cracking of fuel cladding can be adversely affected by the presence of hydrides, especially when they are oriented towards the radial direction of the tubing.

Hydride precipitation is a complicated function of the solubility of hydrogen in cladding materials, cladding hydrogen concentration, stress state, temperature, cooling rate and thermal cycling. Hydride reorientation involves the dissolution of circumferential hydrides and the formation of radial hydrides [7, 8]. The hydrides, present in the circumferential planes of the cladding tubes, transform to a radial orientation when precipitation occurs under loading. For the cladding tubes having the radial hydrides, the microcracks may form at the radial hydrides when the tensile hoop stress exceeds the critical stress for the microcrack formation. These microcracks may link up with the increase of the tensile hoop stress and then propagate along the radial hydrides. This microcrack propagation may continue through the interlinked radial hydride, which generates the brittle fracture [9].

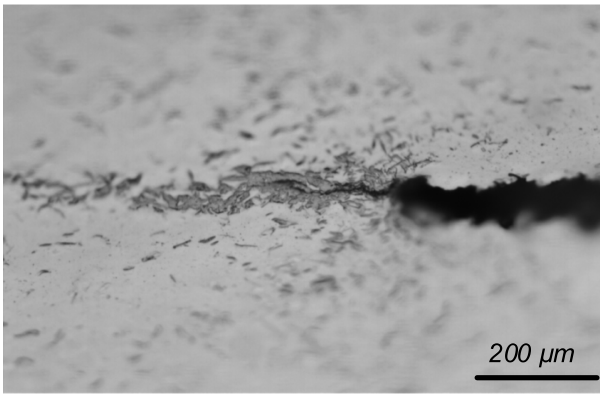

Fig. 1. Hydride crack in RBMK Zr-1Nb cladding specimen. Test temperature 250 °C

These phenomena may strongly influence the mechanical properties of structural materials used in nuclear power plants. The areas of possible hydride reorientation are tube defects with increased stress zones. Characteristic cladding defects are nodular corrosion, stress corrosion or delayed hydride cracking. For instance, the studies [10]

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show that at a burnup of 1.3 MW·d/kgU the surface of a RBMK-1000 fuel rod can develop nodules up to 40 µm deep (outside) and up to 150 µm deep (inner) under the spacer grids. As fuel burnup increases to 19.3 MW·d/kg U, these values increase to 130 µm and 380 µm, respectively. Some fuel claddings, adjacent to the spacer grids, show signs of localised thinning caused by fretting-corrosion. The thinning reaches 400 µm in depth. The appearance of these fuel rods is shown in Fig. 2. The given nodules' depth and fretting-wear (reaching 400 µm) may substantially limit the mechanical strength of a cladding. Crack formation due to internal pressure is possible when such surface defects are formed during operation.

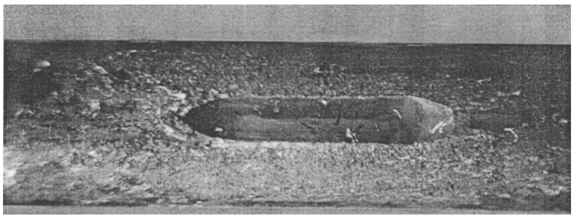

Fig. 2. Appearance of the surface flaw of fuel rods in the spacer grid region [10]

After operation cladding material further undergoes structural changes under impact of temperature and stress during the long term storage period. Some regions of the cladding have higher temperature, over a long period of time hydrogen can migrate from the hotter regions to the colder ones or from zones with lower stress to a higher stress zones. In these regions can form spots of reoriented hydrides with local hydrogen concentration up to several hundred or thousand ppm. Illustration of such hydride accumulation zones is shown in Fig. 3 [11].

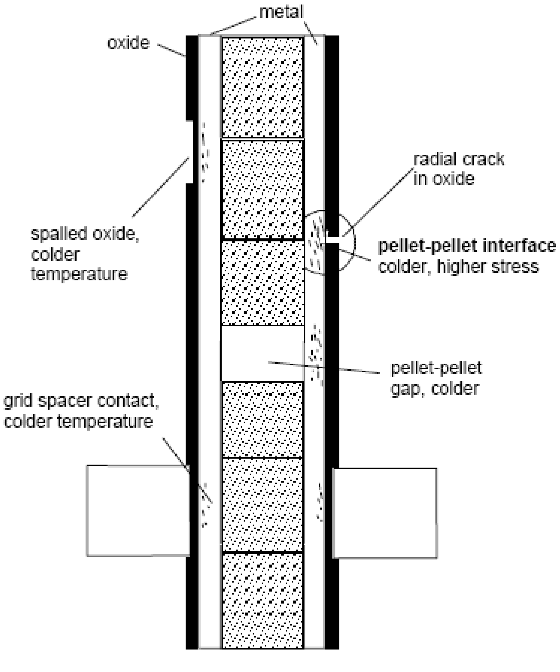

Fig. 3. Illustration of formation potential hydrogen accumulation spots and hydride blisters in spent fuel cladding [11]

Hydride blister formation was found in some high burn up fuel cladding where higher stresses were due to fuel pellet expansion and interaction with cladding wall, as well as at lower temperature zones. These cases were found for high burnup Zry-4 PWR fuel cladding (>55 MWd/kgU) [12, 13].

Several methods are known for testing of thin-walled cladding tubes: centre-cracked half-tube loaded in tension [5], a centre-cracked length of tube loaded by a wedge and mandrel (SPLIT test) [14] and the Pin-Loading Tension (PLT) technique [15]. In some works [7] cladding tube is subjected to thermal cycling in an autoclave under a constant hoop stress by regulating the differential pressure between its internal and external pressures with a constant differential pressure control system.

The material for this study was cold-worked Zircaloy-4 cladding tube. The specimens were made from a hydrided cladding tube having triangular notch that is similar to the surface flaw that may possibly form during operation. The goal of this research was to find out if reorientation can influence cladding failure and determine conditions and possible mechanism of failure.

2. EXPERIMENTAL

2.1. Materials and specimens

The material was cold-worked Zircaloy-4 cladding tube (CW Zry-4). The properties of cladding tube are presented in Table 1. The loading was applied using pin-loading tension (PLT) method [16] and fixture (Fig. 4).

Table 1. Zry-4 cladding tube properties [16]

Manufacturer	Sandvik Steel AB
Batch No.	83786
Thermomechanical treatment	Cold worked
Annealing conditions	Not annealed
Outer diameter (mm)	9.52
Wall thickness (mm)	0.595
Tensile strength (20 °C – 25 °C), N/mm^2	800 – 825
Tensile strength (N/mm²), C 380 – 385 °C	475 – 482

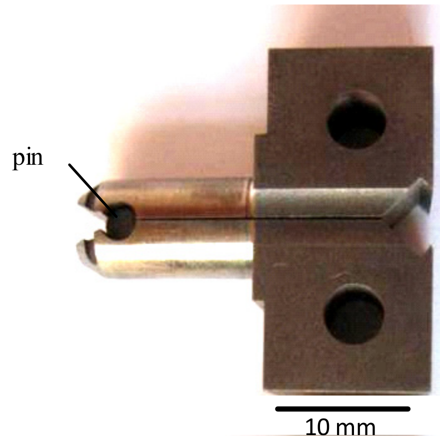

Fig. 4. Pin-loading tension (PLT) fixture

Hydrided cladding material used for the tests was prepared using electrolytic hydriding – diffusion annealing treatment [17]. Cladding tube pieces of 220 mm length were electrolytically hydrided in 0.2M H_2SO_4 at 70 °C using current density 1.5 kA/m^2. Homogenization treatment was performed at 410 °C for 24 hours to get a nominal concentration of about 200 wt-ppm. To reveal microstructure of the tested material, etching composition of $H_2SO_4 : HNO_3 : HF : H_2O$ (3 : 3 : 1 : 3) was used.

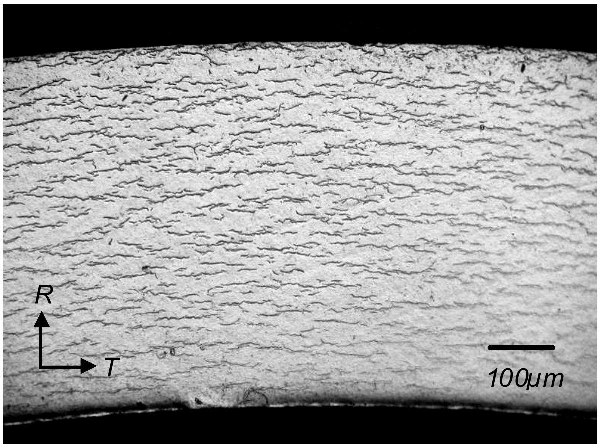

Fig. 5. Hydride microstructure of the tested Zry-4 cladding tube (radial-transverse section)

Metallography of hydride structure on radial-axial and radial-transverse sections shows a uniform hydride distribution with hydrides elongated in the longitudinal direction (Fig. 5).

2.2. Hydride reorientation testing

For hydride reorientation testing stresses in cladding tube usually are created by applying internal pressure [7, 6]. Such method to establish threshold stress for hydride reorientation requires sophisticated equipment. Tests are performed gradually decreasing pressure and using separate specimens until threshold stress value is found. Such long term experiments require not only complex equipment but also considerable time. Described in this work hydride reorientation tests were performed using PLT fixture, but the test specimens were different. For reorientation tests 11 mm length cladding tube specimens were prepared, having two triangular notches along the specimen axis. The measured notch depth is recorded in Table 2. Triangular notch was made similar to the surface flaw that can form during operation (see Fig. 1) and because of increased stress at the notch tip hydride reorientation is possible.

When applying load P on PLT fixture having 11 mm length cladding tube specimen (Fig. 6), load (within the elastic deformation) at a cross section of specimen tube wall will give stress proportional to a distance from the specimen front. The stress will decrease from the maximum value σ_{max} to zero at the specimen end (corresponding to location of fixture axis). In this case stress at the one side of cladding wall will be:

$$\sigma_i = \frac{2P \cdot W \cdot l_x}{l_b^3 \cdot t_i}. \qquad (1)$$

Maximum stresses at the specimen front, when $l_x = l_b$:

$$\sigma_{max} = \frac{2P \cdot W}{l_b^2 \cdot t_i}, \qquad (2)$$

where t_i is the cladding wall thickness (mm) at the given cross section; W is the width of the specimen-fixture assembly (distance between the loading line and the rotation axis), equal 19 mm; l_b is the distance from the rotation axis to the specimen front, equal 11 mm; $l_x = l_b - z$ – distance from the fixture axis at the given cross section, where z is the distance of the given cross section from the specimen front.

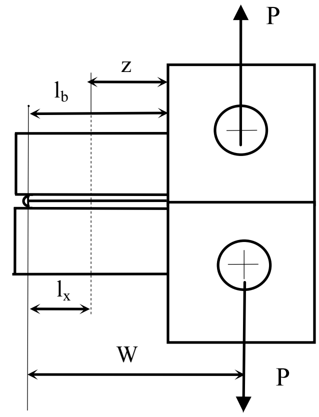

Fig. 6. Specimen – fixture assembly for reorientation testing: W – width of the specimen-fixture assembly (distance between the loading line and the rotation axis), equal 19 mm; l_b – distance from the rotation axis to the specimen front, equal 11 mm; $l_x = l_b - z$ – distance from the fixture axis at the given cross section, where z is distance of the given cross section from the specimen front

Hydride reorientation was performed by cooling the specimen from 340 °C to 40 °C at a cooling rate 0.25 ºC/min. Total duration of the test was 20 h. After testing at the assigned temperature and loading conditions, specimen was examined layer by layer metallographically to determine hydride reorientation under certain length value l_i when hydride reorientation disappears. Knowing this distance, the threshold stress value for hydride reorientation is calculated by equation (1).

3. RESULTS AND DISCUSSION

When the specimen is loaded at a constant temperature, hydride precipitation conditions from the solid solution are different compared to those when temperature is constantly decreasing. When the specimen is cooled from 340 °C without loading, hydride distribution is determined by cladding texture. As the specimen is loaded, higher stresses cause hydride formation near the flaw, perpendicular to the stress direction. In this case orientation of hydrides, which were formed in the bulk of material during the cooling does not change, if no plastic deformation and texture changes take place.

With reference to [18] δ-hydride fracture toughness is very low – 1 MPa·m$^{1/2}$ at 23 °C and (1 – 4) MPa·m$^{1/2}$ at 300 °C. It is claimed that hydride blister, which also consists of δ-hydride has similar fracture toughness value. Applying these threshold stress intensity values it was calculated [19] that critical hydride fracture length is from 11 μm to 102 μm, but up to 150 MPa stress is required for the fracture. Such hydride reorientation can cause brittle fracture or delayed hydride cracking (DHC). For a DHC process condition $K_I \geq K_{IH}$ is required. Threshold value K_{IH} for DHC for CW Zry-4, is about ~5.5 MPa·m$^{1/2}$ [11]. In our tests K_{IH} value was fond (4 – 5) MPa·m$^{1/2}$.

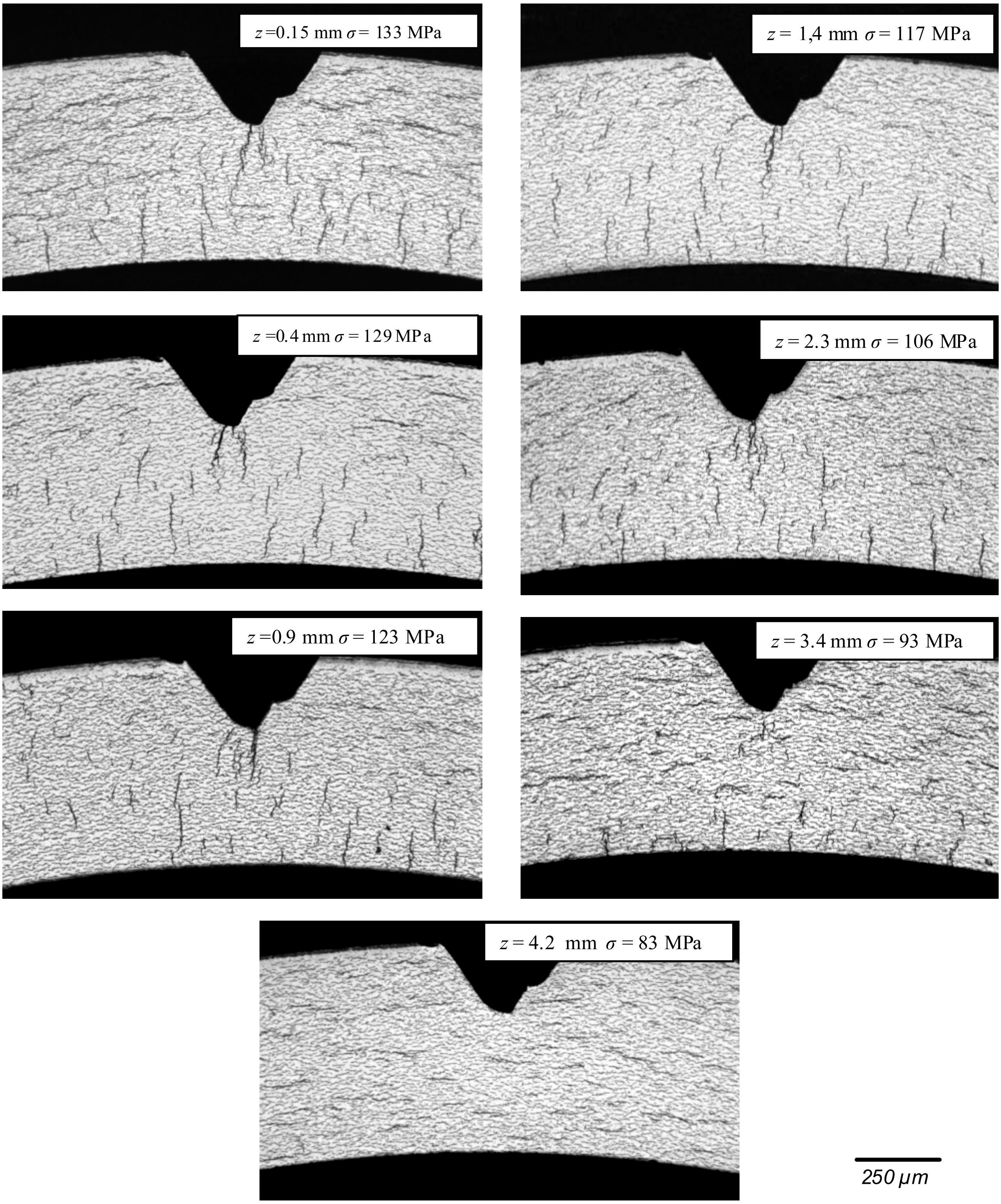

Fig. 7. Hydride reorientation in CW Zry-4 cladding having longitudinal 0.215 mm notch at a different distance *z* from the specimen front. Specimen No 5.1_1

The approximate stress intensity value at the notch could be evaluated by equation [20]:

$$K_I = (\pi a)^{0.5}(\sigma_m f_m + \sigma_b f_b), \qquad (3)$$

Where σ_m ir σ_b are tangential and bending stresses, f_m and f_b crack shape parameters, a is the crack depth. According to [20], for cylindrical tube with a long surface crack, the conditions $a/t \leq 0.8$ and $0.1 \leq t/R_i \leq 0.25$ (t is the cladding tube thickness; R_i is the inner radius), should be satisfied, then $f_m = 1.68$. In this case, we assume that bending stress $\sigma_b \approx 0$ is low compared to the tangential stress estimating that notch tip is near the wall centre and cladding wall is thin.

Presented data show that two fracture modes are possible: brittle fracture caused by hydride reorientation or fracture because of initiated DHC.

Hydride distribution in the cladding wall cross-sections at a different distance from the specimen front (Fig. 6) is shown in Fig. 7. The hydride reorientation primarily occurs at the notch tip. It is obvious that the length of hydrides is comparable to the total length of the cladding wall thickness. Hydride reorientation happens when stress reaches 77 MPa – 89 MPa (Table 2). At a lower stress values hydride location remains unchanged. The obtained threshold reorientation value is similar to the research [21] in case of recrystallized Zircaloy-2 sheet, but is higher than that determined for irradiated BWR cladding [6].

The loading used for reorientation test of the two first specimens did not cause fracture, although reoriented hydride length was up to 200 μm at 180 MPa stress.

Table 2. Hydride reorientation conditions in Zry-4 CW cladding

ID No.	Notch depth a, mm	σ_{max}, MPa	K_{Imax}, MPa·m$^{0.5}$	Threshold conditions for hydride reorientation		
		$l_x = 0$		Cross-section location l_{xi}–l_{xi+1}, mm	Stress range σ_i–σ_{i+1}, MPa	Average stress σ_v, MPa
5.1-1	0.215	134	3.7	6.8 – 7.6	83 – 93	88
	0.12	107	2.8	7.8 – 8.8	76 – 86	81
5.1-4	0.178	180	5.0	5.0 – 5.5	82 – 90	86
	0.098	151	3.7	5.0 – 6.3	69 – 87	73
5.1-5	0.19	218	6.2	–	–	–
	0.19					
Mean					77 – 89	82

As it is shown in Table 2, $K_{Imax} \leq K_{IH}$ that is K_{Imax} values could be too low to initiate DHC. Therefore for the test of specimen No 5.1.5 the load was increased to $K_{Imax} = 6.2$ MPa·m$^{1/2}$. It was supposed that without fatigue crack DHC would not start or testing time will be long. Therefore test was performed at a constant temperature 250 °C after cooling from 340 °C. Nevertheless, as it is shown in Fig. 8 after 3 h of incubation time DHC growth started and in the next 3 h failure of the cladding occurred. The DHC growth is clearly visible at the fracture surface (Fig. 9).

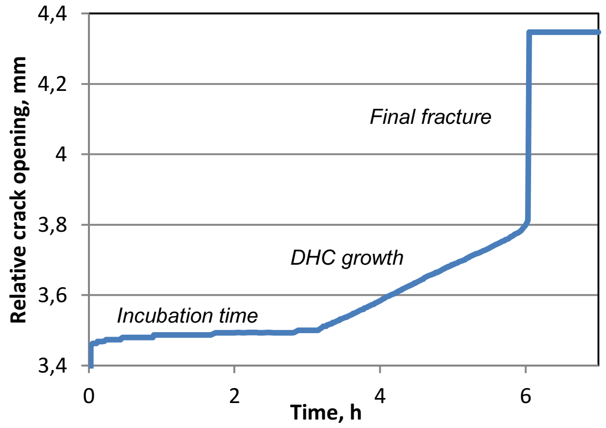

Fig. 8. Testing of Zry-4 CW cladding at 250 °C: relative crack opening vs. time curve

Fig. 9. Fracture surface of Zry-4 CW cladding after testing at 250 °C: 1 – DHC crack; 2 – brittle fracture surface; 3 – notch (3). Specimen ID 5.1_5

Measured DHC crack length was 3.5 mm. As we can observe from the fractograph, crack growth initially started at some crack length, which was less than 3.5 mm as long as a condition $K_I \geq K_{IH}$ was valid. While crack opening continued, K_I increased and crack was growing in both radial and axial directions. Brittle fracture occurred when threshold stress was reached as wall thickness has reduced because of DHC crack.

4. CONCLUSIONS

In case of a surface flaw the reorientation of hydrides takes place perpendicular to the stress direction. The hydride reorientation occurs at a stress range 77 MPa – 89 MPa. At experimental conditions hydride reorientation had not caused fracture at a stress up to 218 MPa. Fracture at the notch tip was initiated by DHC mechanism at K_I value 6.2 MPa·m$^{1/2}$ when a condition for DHC initiation $K_I > K_{IH}$ was fulfilled. For initiation of DHC process larger stress is required than for hydride reorientation. However brittle fracture was caused after some critical DHC crack depth in fuel cladding wall was reached.

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