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Effect of Hydrogen, Hydride Orientation and Temperature on Low-Cycle Fatigue Resistance of Zr-1%Nb Fuel Rod Claddings

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Low-cycle fatigue testing was conducted on annular samples with an outer diameter of 9.13 mm, a wall thickness of 0.68 mm and a width of 2.7 mm, namely: non-hydrogenated samples (cut out of standard Zr-1%Nb cladding tubes); hydrogenated samples with a hydrogen concentration of 50 ... 400 ppm; samples cut out from hydrogenated dummy claddings after hydride reorientation tests performed according to various test modes. The tests were conducted at the temperatures of 25, 180, 350, 400 and 450 °C. The results obtained demonstrate that with increasing the hydrogen content in Zr-1%Nb alloy claddings the fatigue life increases.

Keywords: hydrogen, hydrogen embrittlement, hydrides, hydride reorientation, Zr-1%Nb alloy, «dry» storage, fatigue.

Introduction

During reactor operation, fuel rod cladding is exposed to certain impacts, including vibration loads, internal pressure of gaseous fission products, fuel swelling pressure, thermal stresses in cladding, fuel-cladding frictional force, coolant pressure. Periodically recurring loads also take place during subsequent SNF handling [1]. Based on the function (nuclear fuel protection both during reactor operation and while SNF handling) and the risk of consequences of fuel rod leakage, a number of requirements are specified for the cladding, including the fatigue strength requirements: the strength criterion SC4 (fatigue strength and long-term cladding strength) [2]. During reactor operation, zirconium fuel rod claddings accumulate hydrogen, which is one of the main performance degradation factors during further SNF handling. The effect of hydrogen on mechanical properties depends on many factors: its state (solid solution or hydrides), hydride orientation (tangential or radial), testing temperature etc. 3]. These factors determined the testing and investigations performed, the results of which are provided in this paper.

Materials and Methods

The samples for the fatigue tests were ∅ 9.13× 0.68 mm rings, 2.7 mm wide with a slit width corresponding to the transverse strain value. The low-cycle fatigue tests (LCF) were carried out by transverse deformation of C-shaped samples (cylinders cut along the axis) at the temperatures of 25, 180, 350, 400 and 450 °C. The criterion for assessing the low-cycle fatigue resistance of the samples was the number of cycles to failure. A facility diagram, a method used for low-cycle fatigue testing, an electromechanical circuit for automatic recording the number of cycles to failure, and fatigue test samples are provided in [4], [5]. The frequency of cycles was ~ 0.2 Hz. The majority of tests were conducted at the strain amplitude of 9.75×10^{-3}.

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The set of tests and investigations performed included:

- metallographic studies of hydride morphology in the samples prepared for low-cycle fatigue testing;
- conduct of low-cycle fatigue testing;
- fracture pattern study (point of fracture imaging at 50× magnification) and analyzing images of samples with various hydrogen content after low-cycle fatigue testing;
- investigation of hydride morphology near fracture (performed on all samples subjected to low-cycle fatigue testing).

The fatigue testing was performed on annular samples with an outer diameter of 9.13 mm, a wall thickness of 0.68 mm and a width of 2.7 mm:

- samples cut out from Zr-1%Nb cladding tube in as-received state;
- hydrogenated samples (with a hydrogen content of 50, 100, 200, 300 and 400 ppm)
- hydrogenated samples (up to 300 ppm), cut out from Zr-1%Nb fuel rod dummies under internal pressure (^{293}P = 5 MPa), subjected to hydride reorientation tests in the modes:

heating to 410 °C at the rate of 10 °C/min → holding for 8 hours → cooling at the rate of ~ 2…4 °C/min (mode 1);

heating to 410 °C at the rate of 10 °C/min → holding for 3 hours → cooling to 300 °C at the rate of ~ 2…4 °C/min and 3 subsequent thermal cycles with holding for 1.5 hours at 410 °C and for 1 hour at 300 °C (mode 2);

heating to 410 °C at the rate of 10 °C/min → holding for 3 hours → cooling to 180 °C at the rate of ~ 2…4 °C/min and 3 subsequent thermal cycles with holding for 1.5 hours at 180 °C (mode 3).

Tables 1-5 present the results of LCF tests of non-hydrogenated annular samples cut out from a Zr-1 %Nb cladding tube in as-received state and tested at the temperatures of 25, 180, 350, 400, 450 °C, oscillation frequency of ~ 0,2 Hz and strain amplitudes of 1.3×10^{-2} – 9.75×10^{-3}.

Table 1 – Number of cycles to failure (N) of non-hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 25 °C. Oscillation frequency: 0.2 Hz

Sample No	Strain amplitude	Number of cycles
1	1.3×10^{-2}	1442
2	1.3×10^{-2}	1556
3	1×10^{-2}	4088
4	9.75×10^{-3}	4668
18	9.75×10^{-3}	4798
30	9.75×10^{-3}	5300

Table 2 – Number of cycles to failure (N) of non-hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 180 °C. Oscillation frequency: 0.2 Hz

Sample No	Strain amplitude	Number of cycles
5	1.3×10^{-2}	1448
7	9.75×10^{-3}	3968
8	9.75×10^{-3}	3961

Table 3 – Number of cycles to failure (N) of non-hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 350 °C. Oscillation frequency: 0.2 Hz

Sample No	Strain amplitude	Number of cycles
14	1.3×10^{-2}	1006
15	1.3×10^{-2}	1230
13	1.3×10^{-2}	1298
10	1×10^{-2}	2500
17	9.75×10^{-3}	2750
12	7.5×10^{-3}	8500

Table 4 – Number of cycles to failure (N) of non-hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 400 °C. Oscillation frequency: 0.2 Hz

Sample No	Strain amplitude	Number of cycles
49	9.75×10^{-3}	1895
74	9.75×10^{-3}	2697
76	9.75×10^{-3}	2160

Table 5 – Number of cycles to failure (N) of non-hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 450 °C. Oscillation frequency: 0.2 Hz

Sample No	Strain amplitude	Number of cycles
54	9.75×10^{-3}	1905
55	9.75×10^{-3}	1105
75	9.75×10^{-3}	1980

The results obtained demonstrate that with increasing the temperature, the number of cycles to failure decreases. This result indicates an insignificant decrease in the ductile properties of the material at the oscillation frequency of 0.2 Hz and the strain amplitudes of 1.3×10^{-2} – 9.75×10^{-3}.

Tables 6-10 provide the results of LCF tests of hydrogenated annular samples (with the hydrogen content of 50, 100, 200, 300 and 400 ppm) cut out of Zr-1%Nb fuel rod claddings and tested at the temperatures of 25, 180, 350, 400, 450°C, oscillation frequency of 0.2 Hz and strain amplitude of 9.75×10^{-3}.

Table 6 – Number of cycles to failure (N) of hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 20 °C. Oscillation frequency: 0.2 Hz

Sample No	[H]tppm	Strain amplitude	Number of cycles
19	50	9.75×10^{-3}	4360
20	50	9.75×10^{-3}	5760
21	100	9.75×10^{-3}	7563
22	100	9.75×10^{-3}	7741
23	200	9.75×10^{-3}	6220
24	200	9.75×10^{-3}	7920
26	300	9.75×10^{-3}	6840
27	400	9.75×10^{-3}	11400
28	400	9.75×10^{-3}	7920

Table 7 – Number of cycles to failure (N) of hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 180 °C. Oscillation frequency: 0.2 Hz

Sample No	[H] tppm	Strain amplitude	Number of cycles
42	300	9.75×10^{-3}	5430
33	300	9.75×10^{-3}	6702

Table 8 – Number of cycles to failure (N) of hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 350 °C. Oscillation frequency: 0.2 Hz

Sample No	[H]tppm	Strain amplitude	Number of cycles
17	50	9.75×10^{-3}	3240
12	100	9.75×10^{-3}	3405
13	200	9.75×10^{-3}	3680
14	300	9.75×10^{-3}	3240
15	400	9.75×10^{-3}	4320
9	200	1.0×10^{-3}	3200
11	200	7.5×10^{-3}	10100
43	300	9.75×10^{-3}	3540

Table 9 – Number of cycles to failure (N) of hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 400 °C. Oscillation frequency: 0.2 Hz

Sample No	[H]tppm	Strain amplitude	Number of cycles
70	50	9.75×10^{-3}	3400
71	100	9.75×10^{-3}	3210
72	300	9.75×10^{-3}	3240
73	400	9.75×10^{-3}	3288

Table 10 – Number of cycles to failure (N) of hydrogenated Zr-1%Nb cladding tube samples during fatigue testing at 450 °C. Oscillation frequency: 0.2 Hz

Sample No	[H]tppm	Strain amplitude	Number of cycles
56	50	9.75×10^{-3}	2600
57	100	9.75×10^{-3}	2200
58	200	9.75×10^{-3}	2340
59	300	9.75×10^{-3}	2520
60	400	9.75×10^{-3}	2460

The results obtained demonstrate that at room temperature, the fatigue resistance increases with increasing the hydrogen content up to 400 ppm, while at the temperatures above 350 °C, the fatigue resistance decreases.

Tables 11-13 show the results of LCF tests conducted on annular samples cut out from hydrogenated (300 ppm) dummy claddings tested under internal pressure (P293=5 Pa) in the modes 1-3.

Analysis of the data given in tables 1-10 shows:

- with increasing the test temperature, the number of cycles to failure decreases;
- with increasing the strain amplitude, the number of cycles to failure decreases;
- at the temperatures of 350, 400, 450 °C, the fatigue resistance of hydrogenated samples slightly increases as compared to non-hydrogenated ones (Figure 1);
- at room temperature and at 180 °C, the fatigue resistance of Zr-1%Nb claddings increases with increasing the hydrogen content.

Analysis of the data from Tables 1 – 10 and Tables 11-13 demonstrate that:

- a significant hydride reorientation, which occurs in the claddings with increasing the hydrogen content up to 400ppm, does not lead to a decrease in the fatigue resistance at 25, 180 and 350 °C (Figure 2).

Figures 3-4 show typical results of metallographic studies of cross sections of dummy claddings, damaged samples in the area adjacent to the fracture surface, along with the results for the same samples before fracture.

Figure 3 shows a hydride orientation typical of the dummy claddings before the hydride reorientation test (tangential, set by the manufacturing technology).

Figure 4 shows reoriented hydrides cut out from hydrogenated (400 ppm) cladding of dummy No 48 tested for hydride reorientation in the mode 2.

Table 11 – Number of cycles to failure during low-cycle fatigue testing of samples cut out of dummy claddings with a hydrogen content of 300 ppm tested in the mode 1

Sample No	$Fn_{\text{before test}}$	T_{test} °C	Strain amplitude	No of cycles	$Fn_{\text{after test}}$
35	0.68	20	9.75×10^{-3}	9360	0.42
36	0.74	20	9.75×10^{-3}	10500	0.46
38	0.7	350	9.75×10^{-3}	2520	0.41

Table 12 – Number of cycles to failure during low-cycle fatigue testing of samples cut out of dummy claddings with a hydrogen content of 300 ppm tested in the mode 2

Sample No	$Fn_{\text{before test}}$	T_{test} °C	Strain amplitude	No of cycles	$Fn_{\text{after test}}$
39	0.67	20	9.75×10^{-3}	10400	0.5
41	0.86	180	9.75×10^{-3}	5380	0.48
42	0.74	350	9.75×10^{-3}	3380	0.52

Table 13 – Number of cycles to failure during low-cycle fatigue testing of samples cut out of dummy claddings with a hydrogen content of 300 ppm tested in the mode 3

Sample No	$Fn_{\text{before test}}$	T_{test} °C	Strain amplitude	No of cycles	$Fn_{\text{after test}}$
46	0.92	20	9.75×10^{-3}	10080	0.49
45	0.94	180	9.75×10^{-3}	5510	0.57
49	0.78	20	9.75×10^{-3}	10400	0.65
47	0.82	180	9.75×10^{-3}	6100	0.49
48	0.96	350	9.75×10^{-3}	3240	0.53

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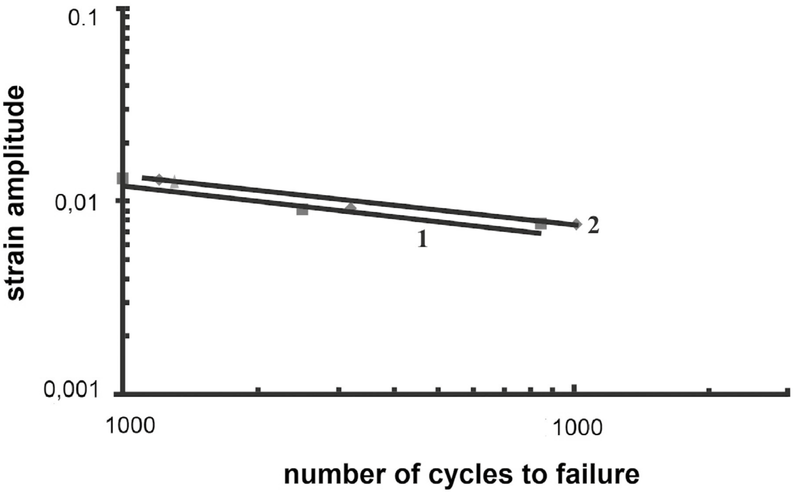

Figure 1 – Fatigue curves of Zr – 1 % Nb cladding tube material at 350 °C: ■– Zr-1%Nb alloy, ♦– Zr-1%Nb alloy with a hydrogen content of 200 ppm

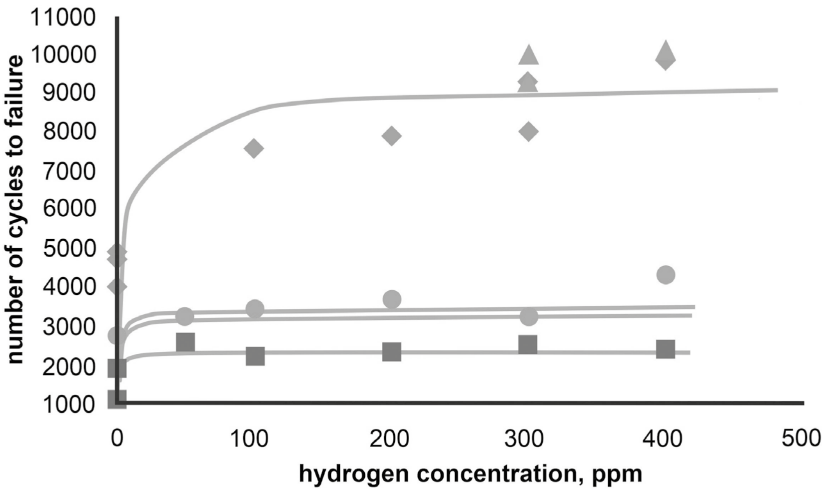

Figure 2 – Number of cycles to failure N_c dependence on temperature: ♦ – 20, ●– 350,+ – 400, □– 450 °C) and hydrogen content (0 – 400 ppm). Reoriented samples ▲ – No 35, No 36, No 46, No 49,✱ – No 42, No 48

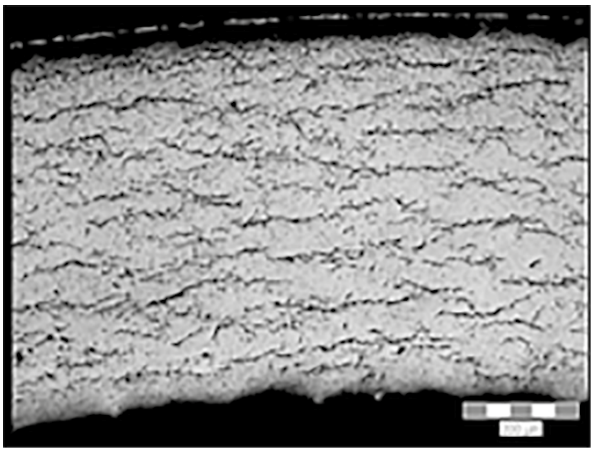

Figure 3 – Hydrides in Zr-1%Nb claddings with hydrogen content of 200 ppm, Fn = 0.08. Hydride morphology typical of hydrogenated dummy claddings

Figure 4 – Sample No 48: 400 ppm, P 293=5.0 MPa, 3 thermal cycles 410↔300 °C, Fn≈ 0.96

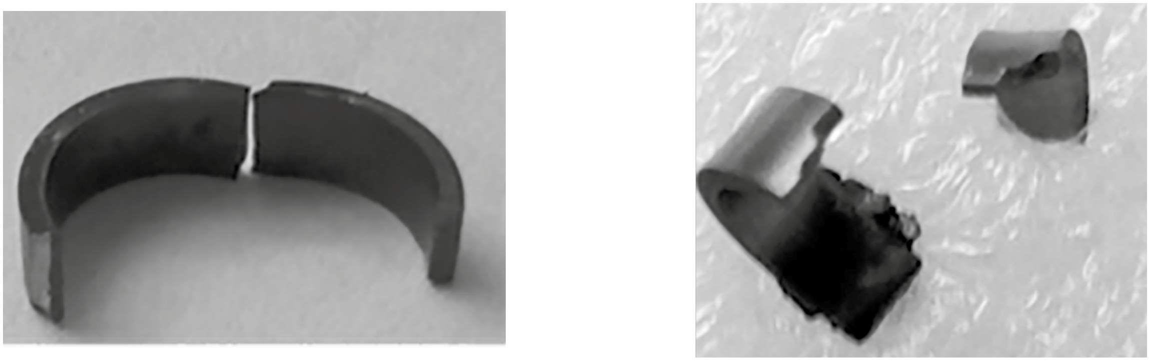

Figure 5 – A sample with a hydrogen content of 400 ppm damaged during fatigue testing (oscillation frequency: 0.2 Hz, strain amplitude: 9.75×10^{-3}). Number of cycles to failure: 10410. A typical example of ductile fracture

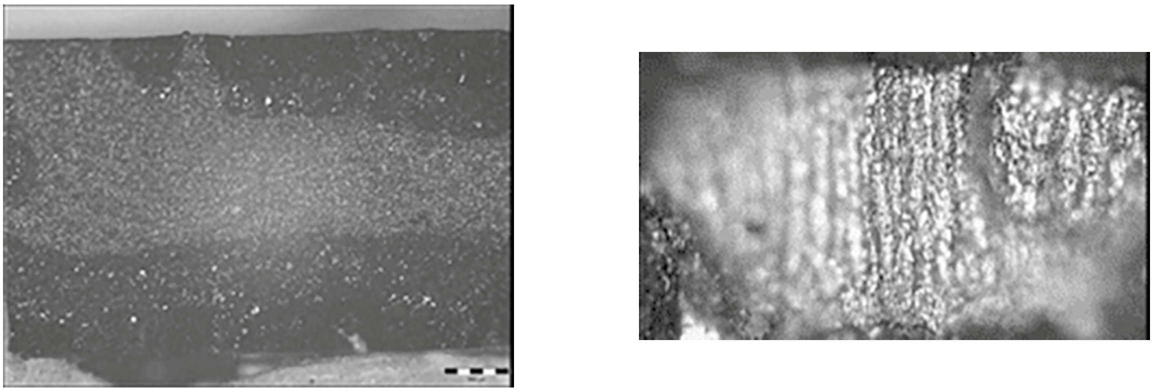

Figure 6 – Ductile fracture. A sample with hydrogen content of 400 ppm damaged during fatigue testing (oscillation frequency: 0.2 Hz, strain amplitude: 9.75×10^{-3}). Number of cycles to failure: 10410

Figures 5 and 6 show typical ductile fracture after LCF testing.

As can be seen from the comparison of hydride morphology in Figures 3-4, the tests conducted in the modes used in this work lead to a significant hydride reorientation over the entire dummy fuel rod cladding, as well as to changing the hydride orientation coefficient in the area adjacent to the fracture surface.

It has to be noted that after the fatigue tests, the hydride orientation coefficient ranges within 0.5 – 0.6. There is the reason to believe that during the fatigue tests (and most likely, during other types of strain exposures), a "strain-induced" hydride reorientation takes place, which is of considerable independent interest, both fundamental and practical. For a better understanding, there is a need for a focused and detailed study.

Based on all results obtained for the samples cut out from the claddings with various hydrogen content of 50…400 ppm tested for hydride reorientation in the modes simulating handling operations with a limiting heating up to 410 °C and accidents with 3 thermal cycles 300 ↔ 410 °C (180 ↔ 410 °C) further subjected to the low cycle fatigue testing at 20, 180, 350 °C with various number of cycles, we can state with the confidence that all tested claddings underwent ductile fracture, which confirms retention of the ductile properties of the material.

Conclusion

The effect of hydrogen and hydride orientation on the low-cycle fatigue resistance of Zr-1%Nb cladding tubes at 20, 180, 350, 400, 450 °C at a hydrogen content increased to 400 ppm was investigated. Based on the investigation results it can be concluded that:

- hydrogenation of Zr-1%Nb claddings increases their fatigue life which may be associated with increasing the material strength;
- hydride reorientation does not affect the fatigue resistance of Zr-1%Nb claddings;
- the fatigue strength increases with decreasing the test temperature, which is in agreement with the results of [6].

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Вплив водню, орієнтації гідридів та температури на опір малоциклової втоми оболонок твелів зі сплаву Zr-1%Nb

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Умови навантаження оболонок твелів під час поводження з відпрацьованим ядерним паливом, різноманітність механізмів їх деградації дуже ускладнюють діагностику їх характеру пошкоджуваності і руйнування. Різні види напруженого стану з урахуванням структурного стану матеріалів, міцності і пластичних властивостей у процесі призводять до різних видів дефектів. Також велику роль відіграє водень, присутній в оболонках твела відпрацьованого ядерного палива, як у вигляді водню, так і у вигляді гідридів.

Опір малоцикловій втомі є однією з основних характеристик міцності конструкційних матеріалів, що зазнають під час експлуатації циклічних навантажень. Характеристики малоциклової втоми затребувані під час розрахунків на міцність і ресурс, а також під час порівняння подібних матеріалів. Випробування на малоциклову втому здійснюються в пластичній області, тобто під час навантажень або деформацій, що перевищують межу плинності матеріалу. Керувати і контролювати процес випробувань можна, використовуючи один з двох параметрів – напруження або деформації. Актуальним є дослідження і визначення характеристик малоциклової втоми за заданої деформації. Отримані до нинішнього часу дані з цього питання вельми обмежені і належать переважно до сплавів циркалой.

У цій статті наведено випробування на малоциклову втому на кільцевих зразках зі сплаву Zr-1%Nb із зовнішнім діаметром 9,13 мм, товщиною стінки 0,68 мм і шириною 2,7 мм: негідрованих (вирізаних з штатних оболонок труб Zr-1%Nb), гідрованих з вмістом водню 50 ... 400 ppm (вирізаних з гідрованих оболонок макетів після випробувань на переорієнтацію гідридів по різних режимах). Випробування проводилися при температурах 25, 180, 350, 400 і 450 °C. Згідно з отриманими результатами, з підвищенням вмісту водню в оболонці зі сплаву Zr-1%Nb спостерігається підвищення втомної довговічності.

Ключові слова: водень, водневе окрихчення , гідриди, переорієнтація гідридів, сплав Zr-1%Nb, «сухе» сховище, втома.

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