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Journal of Nuclear Materials 280 (2000) 106–110

journal of nuclear materials

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Influence of irradiation on K_{ISCC} of Zr–1%Nb claddings

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Received 10 July 1999; accepted 17 January 2000

Abstract

Experimental results on iodine induced stress corrosion cracking (SCC) in irradiated claddings from Zr–1%Nb alloy are analyzed. Fatigue cracks were grown at their inner surfaces. The irradiation was carried on in the liquid sodium cooled BOR-60 to the fluence not lower than 10^{22} n/cm^2. The SCC-test was carried at 350°C, in argon gas at constant pressure and iodine surface concentration of 0.2 mg/cm^2. The threshold stress intensity factor for the irradiated Zr–1%Nb claddings was determined to be ~2.0 MPa$\sqrt{\text{m}}$.

1. Introduction

Bibilashvili et al. [1] studied the laws governing the crack evolution in unirradiated Zr–1%Nb claddings under stress corrosion cracking (SCC) conditions. The experimental procedure consisted in testing pre-flawed claddings internally pressurized with an inert gas containing iodine at the specified concentration. The tests performed in the studies resulted in establishing the crack velocity vs stress intensity factor relationship and the threshold value of K_{ISCC} for unirradiated Zr–1%Nb alloy.

The implemented investigations enabled us to proceed to the next stage of the work, namely, the assessment of the influence exerted by irradiation on the cladding fracture in SCC. For this purpose cladding samples having a fatigue defect were irradiated in BOR-60 and tested in hot-cell. The paper reviews the results of studying crack velocities in Zr–1%Nb claddings under SCC conditions and discusses the influence of irradiation on the crack resistance of various zirconium alloys.

2. Experimental conditions

2.1. Irradiation

The samples to be irradiated were prepared from Zr–1%Nb alloy claddings and had the following sizes:13.6 mm in diameter, 0.9 mm for wall thickness and 140 mm long. The chemical composition of Zr–1%Nb (E110) cladding alloy and the final heat treatment temperature are given by Nikulina et al. [2]. Fatigue cracks were grown at their inner surfaces. Fatigue pre-cracking was produced in the facility of cyclic loading at the amplitude of ~120 μm and the frequency of 5 Hz. The number of the cycles was ~40 000. The samples were evacuated, filled with Ar at a pressure of 1 kgf/cm^2 and sealed by welding. No iodine was introduced.

The irradiation was carried on in the liquid sodium cooled BOR-60. The objective was to irradiate the claddings to the fluence not lower than 10^{22} n/cm^2 at a temperature close to the testing temperature (350°C). The samples were positioned in an assembly intended for materials science studies at four levels to provide the following irradiation conditions:

- the temperature (325–340)°C;
- the fluence (1.0–1.5) 10^{22} n/cm^2 ($E > 0.1$ MeV).

The design of the experimental assembly did not rule out the wetting of the sample by sodium during irradiation.

2.2. Post-irradiation tests and examinations

The irradiated samples were transferred to a hot-cell where one of the plugs was cut-off and ampoules containing iodine were inserted. Each sample was connected to the gas supply line of a high pressure facility and mechanically sealed (not welded). The sample together

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PII: S0022-3115(00)00012-X

with the sealing washers was placed into a furnace preheated to 350°C. After ~15 min heating controlled by a thermocouple installed on the sample the pressure within it was raised to achieve the specified value during 10–15 s. Starting from this moment the counter was set on. The general schema of the facility is illustrated in Fig. 1.

The tests were carried on until the sample lost its tightness or during a specified period of time. The moment the sample lost its tightness was identified by a pressure drop in the gas supply line.

After the test the cracked surfaces were examined with optical and scanning electron microscopes. The samples were prepared in the following way. A cracked annular section was cut out of the sample. The annulus was broken up along the crack with the resultant two fragments having the crack fracture surfaces. As a rule, the sample was broken up straight before the microscopic examinations to minimize the surface oxidation. Before the electron microscope examinations the fracture surface of some samples was cleaned ultrasonically.

2.3. Calculation of stress intensity factor

The stress intensity factor was calculated from the analytic formula given by Newman and Raju [3] and recommended in the Stress Intensity Factor Handbook [4]. The calculation was performed for the initial defect sizes.

It is to be noted that in [4] for the calculation of the stress intensity factors stresses at the inner surface of a cladding are used. Therefore, the hoop stresses were calculated with the following formula:

$$\sigma_\theta = (P_{\text{test}} D_{\text{in}})/(2\delta) \tag{1}$$

where P_{test} is pressure gauge indications during testing, MPa, D_{in} is the inner diameter of a cladding, and δ is the wall thickness of a cladding. The size of the corrosion induced crack evolution region was assessed from the surface in the area of the maximal crack propagation. The crack velocity was calculated from the size of the SCC area and the overall testing time under the steady-state conditions.

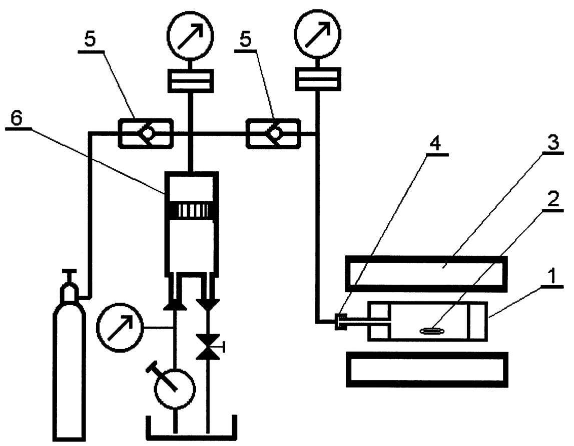

Fig. 1. Schematic description of high pressure testing facility: (1) Sample; (2) ampoule containing iodine; (3) heater; (4) mechanical sealing; (5) isolation valve; (6) pressure booster.

3. Testing results

The results of the test are illustrated in Fig. 2. The studies of the unirradiated claddings reveal that the crack velocity vs stress intensity factor relationship is described by two regions, namely, the drastic velocity acceleration one at $K_{\text{I}} \approx K_{\text{ISCC}}$ and the nearly constant crack velocity one at $K_{\text{I}} > K_{\text{ISCC}}$ [1]. The results of the experiments with the irradiated Zry-4 claddings given by Schuster et al. [5] point to a similar mode of the dependence. The crack velocity vs stress intensity factor plot is shown in Fig. 2 for the irradiated and unirradiated Zr–1%Nb claddings.

It is to be noted that in the samples investigated the crack sizes varied from 380 to 360 μm in the depth and from 2800 to 9900 μm in the length with the hoop stresses being 17–53 MPa. The fracture was not attended with the cladding distortion.

The studied range of K_{I} varied from 0.54 to 7.57 MPa$\sqrt{\text{m}}$. The fracture surfaces after the break-up of the initial defect were studied in some detail at the factors of the stress intensity lower than the threshold value (K_{ISCC}). Thus, at the values of

$K_{\text{I}} = 0.54$ MPa$\sqrt{\text{m}}$ ($\tau = 16$ h);

$K_{\text{I}} = 1.06$ MPa$\sqrt{\text{m}}$ ($\tau = 12$ h);

$K_{\text{I}} = 1.12$ MPa$\sqrt{\text{m}}$ ($\tau = 13$ h);

$K_{\text{I}} = 1.13$ MPa$\sqrt{\text{m}}$ ($\tau = 100$ h);

$K_{\text{I}} = 1.85$ MPa$\sqrt{\text{m}}$ ($\tau = 13$ h),

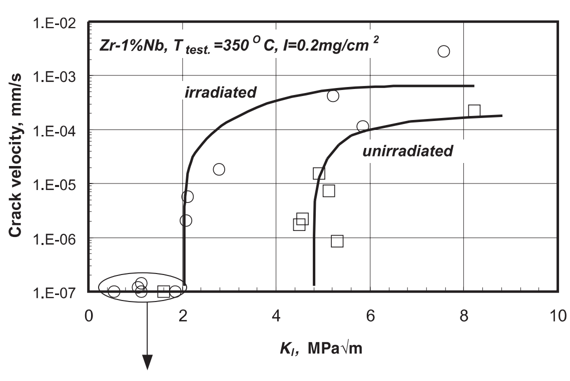

Fig. 2. Average crack velocity ($\Delta a/\Delta t$) vs stress intensity factor (K_{I}) relationship for Zr–1% Nb alloy cladding. $T_{\text{test}} = 350$°C (○ – irradiated Zr–1%Nb, □ – unirradiated Zr–1%Nb).

evidence of the corrosion induced cracking was detected. Fig. 3 illustrates the surface of the fractured sample tested at $K_I = 1.85$ MPa$\sqrt{\text{m}}$ for 13 h.

During this period of time no increment of the defect size took place.

With a further increase of K_I the cracks propagated at the following velocities:

at $K_I = 2.08$ MPa$\sqrt{\text{m}}$, $\Delta a/\Delta t = 2.04 \times 10^{-6}$ mm/s,

at $K_I = 2.11$ MPa$\sqrt{\text{m}}$, $\Delta a/\Delta t = 5.80 \times 10^{-6}$ mm/s,

in other words, with K_I increased by 0.03 MPa$\sqrt{\text{m}}$ the crack velocity becomes a factor of 2.5 higher. The results give an evidence that for the irradiated Zr–1%Nb claddings the threshold stress intensity factor makes up ~2.0 MPa$\sqrt{\text{m}}$. This value is more than twice lower than that of the unirradiated material ($K_{ISCC} = 4.8$ MPa$\sqrt{\text{m}}$) [1]. The fracture surface of the sample is illustrated in Fig. 4. As with the unirradiated sample the inter- and trans-granular types of the fracture are clearly evident.

4. Analysis of the results and discussion

The results acquired give an evidence that under irradiation Zr–1%Nb claddings have a lower threshold stress intensity factor.

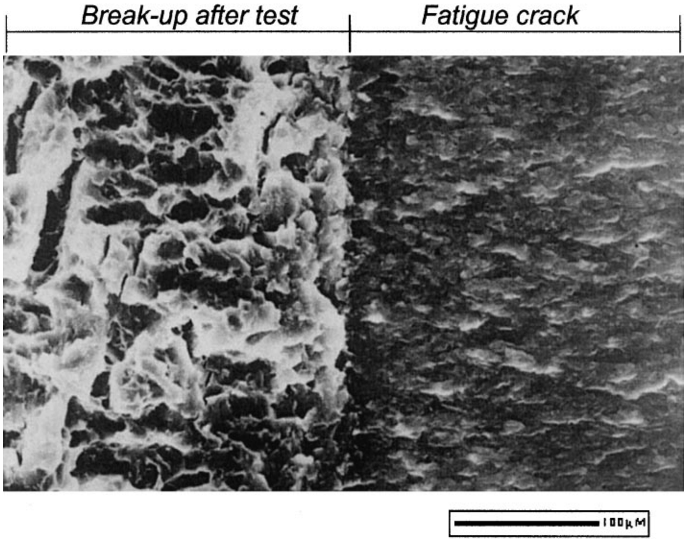

Fig. 3. Fracture surface of irradiated Zr–1%Nb cladding. No SCC is available. $K_I = 1.85$ MPa$\sqrt{\text{m}}$.

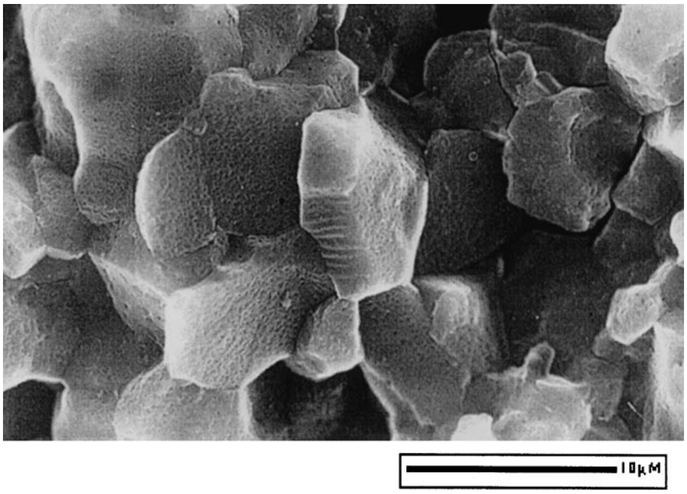

Fig. 4. Fracture surface of irradiated Zr–1%Nb cladding in SCC. $K_I = 2.10$ MPa$\sqrt{\text{m}}$.

Table 1
Conditions of SCC tested material irradiation

Material	Reactor	Reactor coolant	Neutron fluence (n/cm^2)	Irradiation temperature (°C)	Notes
Zr–1%Nb(RX)	BOR-60	Na	1.5×10^{22} ($E > 0.1$ MeV)	325–340	This work
Zircaloy-4(SR)	Siloe′	NaK	1.3×10^{21} ($E > 0.0$ MeV)	–	[5]
Zircaloy-4(SR)	HBR	H_2O	3.6×10^{21} ($E > 0.1$ MeV)	–	[6]
Zircaloy-2(RX)	R2	H_2O	3.0×10^{20} ($E > 1.0$ MeV)	300	[7]

The influence of irradiation on the above mentioned factor is discussed below. Table 1 lists the irradiation conditions reported in the literature.

The review of the papers investigating the influence induced by the irradiation on fracture in the environment containing iodine reveals the common methodological approach to studying this phenomenon, namely, pre-irradiated internal inert gas (containing iodine) pressurized claddings with purposely made defects at their inner surfaces are used for testing. The only distinctions were in the methods of producing fatigue defects and calculating the stress intensity factors. To give rise to a fatigue crack at the inner surface the cladding was subjected to local cyclic loading [1,5]. In the work by Pettersson [7] the fatigue defect was produced at the tip of the pre-machined notch.

Cubicciotti et al. [6] used claddings having only blunt defects for which the radius of the defect tip was bout 50 μm. The experiments carried out with these claddings (Zry-4) did not reveal any fractures close to defects at $K_I = 2.4$–$3.7\ \mathrm{MPa}\sqrt{\mathrm{m}}$ [6]. The fracture close to the defect occurred at $K_I = 7.9\ \mathrm{MPa}\sqrt{\mathrm{m}}$. It is apparent that the available information is insufficient to assess the influence of irradiation on the threshold stress intensity factor for claddings having defects of this type.

$K_{ISCC} = 4\ \mathrm{MPa}\sqrt{\mathrm{m}}$ was determined by Norring et al. [8] for recrystallized Zry-2 unirradiated claddings with fatigue defects. In the reported tests of the samples irradiated in R2 to the fluence of $3.0 \times 10^{20}\ \mathrm{n/cm^2}$, the lower bound of the threshold stress intensity factor was found to be equal to $\sim 3\ \mathrm{MPa}\sqrt{\mathrm{m}}$ [7]. Although this result did not demonstrate a substantial reduction in K_{ISCC} after the fluence of $3 \times 10^{20}\ \mathrm{n/cm^2}$ was reached, nonetheless, it revealed a tendency towards its reduction under irradiation (Fig. 5).

Aside from this work, the most noticeable reduction (more than twice) was found for Zry-4 (SR) claddings irradiated to the fluence of $1.3 \times 10^{21}\ \mathrm{n/cm^2}$ (Fig. 6) in Ref. [5].

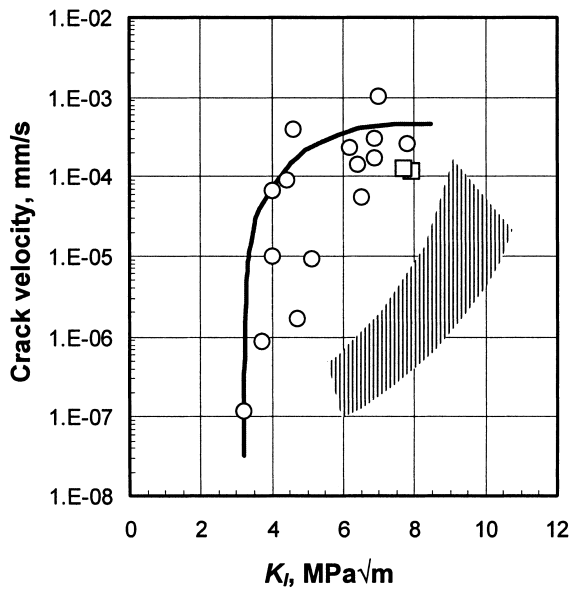

Fig. 5. Influence of irradiation on crack velocity in Zry-2 (RX) claddings. $T_{test} = 320$°C. ○/□ – results on irradiated/unirradiated Zry-2; ▥ – results on unirradiated Zry-2 [7,8].

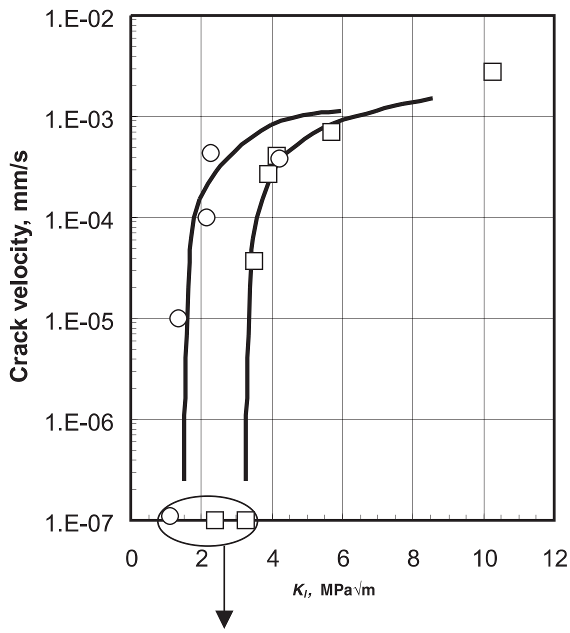

Fig. 6. Influence of irradiation on crack velocity in Zry-4 (SR) claddings. $T_{test} = 350$°C [5]. ○/□ – results on irradiated/unirradiated Zry-4.

Thus, one can assume that the reduction of K_{ISCC} under irradiation is likely to be a common phenomenon typical of zirconium alloys independent of their compositions.

Under irradiation both Zr–1%Nb and other zirconium alloys develop point defects and dislocation loops with the resultant increase of their densities. However, as distinct from Zry-4 no amorphous changes were identified in the Zr–1%Nb alloy. In other words, no amorphization of the β-Nb phase was detected in the work by Shishov et al. [9]. The variations in the microstructure between the irradiated and unirradiated Zr–1%Nb alloy did not substantially affect the fracture mode. The pattern of the fracture surface of the irradiated Zr–1%Nb cladding samples (as tested for SCC) did not reveal any specific features compared to the fracture of unirradiated Zr–1%Nb; the mixed inter- and transgranular fracture is typical of both. However, the fraction of the intergranular fracture in the irradiated material was markedly higher.

The speculation assuming the decohesive effect of iodine on changes in the strength of the zirconium interatomic bonds as a result of the iodine absorption by zirconium is not inconsistent with the facts observed by Cox and Wood [10]. On this assumption the alloy fracture may start at sites where high concentrations of both stress and iodine are available. The tip of a crack or a defect may apparently be such a site. It is experimentally shown by Novikov et al. [11] that iodine in high concentrations is present around the crack tip in the tests under SCC conditions. It is apparent that when in this region the stress exceeds the strength of zirconium reduced by the presence of iodine, a microfracture will initiate with the resultant merging of the microcrack and the main crack. Intergranular cracks are likely to initiate as a result of the microplastic shear deformation along the boundaries that results in the oxide fracture as well as the penetration of iodine and its interaction with zirconium [11,12].

Irradiation may be a factor that reduces the level of external stresses in SCC as a result of the increased density of dislocations and corresponding internal stresses.

5. Conclusion

1. The crack growth of iodine induced SCC was investigated at 350°C using samples of Zr–1%Nb claddings irradiated to a fluence of $(1.0 - 1.2) \times 10^{22}$ n/cm^2.
2. Compared to the unirradiated alloy, the irradiation was shown to reduce the threshold stress intensity factor (K_{ISCC}) from 4.8 to 2.0 MPa$\sqrt{m}$.

Acknowledgements

The authors would like to thank Z. Ostrovskyi (RIAR) for the SEM analysis.

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