Journal of Nuclear Materials 508 (2018) 459–464

Contents lists available at ScienceDirect

Journal of Nuclear Materials

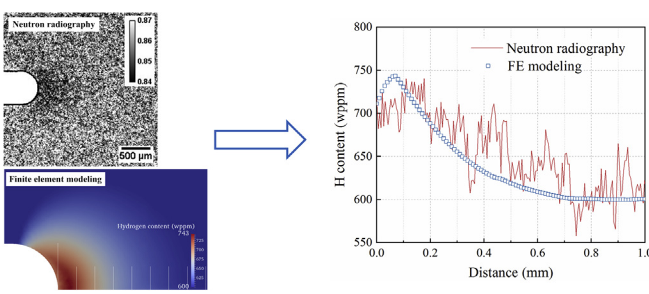
journal homepage: www.elsevier.com/locate/jnucmat

Hydrogen diffusion under stress in Zircaloy: High-resolution neutron radiography and finite element modeling

Weijia Gong [a, *], Pavel Trtik [b], Stéphane Valance [a], Johannes Bertsch [a]

[a] *Laboratory for Nuclear Materials, Paul Scherrer Institut, Villigen PSI, 5232, Switzerland*
[b] *Laboratory for Neutron Scattering and Imaging, Paul Scherrer Institut, Villigen PSI, 5232, Switzerland*

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:
Received 6 December 2017
Received in revised form
4 May 2018
Accepted 29 May 2018
Available online 30 May 2018

Keywords:
Neutron radiography
Zirconium
Hydrogen
Stress-driven diffusion

ABSTRACT

High-resolution neutron radiography (~10 μm) was utilized to quantify hydrogen diffusion under stress in Zircaloy nuclear fuel cladding materials. A notched Zircaloy-4 plate with ~600 wppm hydrogen underwent a thermo-mechanical test to allow hydrogen in solid solution to diffuse solely under a stress raiser. After the test, neutron radiography revealed an elevated concentration field at the notch, with a hydrogen gradient detectable over 700 μm and a maximum elevation of 130 wppm. Finite element computation was then performed by employing the Cauchy stress tensor and hydrogen-induced anisotropic transformation trains as the driving force of hydrogen diffusion.

During operation in a reactor, zirconium based cladding tubes take up a part of the hydrogen produced by the oxidation reaction between the reactor coolant water and the hot rod surface. The absorbed hydrogen, depending on concentration and temperature, in solid solution or as zirconium hydrides can be deleterious in both forms. The hydrides exhibit rather low fracture toughness in comparison with the zirconium matrix. In the presence of hoop stress, circumferential hydrides could be reoriented radially [1,2] weakening the integrity of claddings. In solid solution, hydrogen can diffuse to a stress raiser, precipitate there when the concentration accumulation goes beyond the solubility, followed by hydride growth and eventually provoke a time-dependent mechanism of cracking, so-called delayed hydride cracking (DHC) [3,4]. The hydrogen is therefore the source of degradation mechanisms affecting cladding safe operation, and increasing the risk for the integrity during intermediate dry storage, handling and transportation.

DHC has been investigated with an experimental emphasis on

* Corresponding author.
E-mail address: weijia.gong@psi.ch (W. Gong).

https://doi.org/10.1016/j.jnucmat.2018.05.079

dependence of cracking velocity on microstructure (yield stress) [5], and from the theoretical side thermodynamic descriptions were formulized by Puls [4,6]. However, hydrogen diffusion under stress which is the underlying mechanism of DHC remains open to determine, which can be ascribed to ineffective classical post-mortem characterization. As a rising approach, neutron imaging (either in radiographic or tomographic mode) provides the possibility to quantify hydrogen concentrations field with a concentration resolution of 100 wppm and 30 wppm, respectively, for ex-situ and in-situ measurements [7,8]. It was interesting that the concentration resolution of the technique was recently reported to reach as sensitive as ~5 wppm by optimizing the experimental arrangement and data analysis methodology [9]. However, the spatial resolution has been long limited to several tens of micrometers [8,10–12] which seems to be the last obstacle to reveal localized hydrogen inhomogeneity, for instance, at crack tip vicinity.

In this study, we present quantitative results of stress-induced hydrogen redistribution measured by the cutting-edge detector of PSI Neutron Microscope [13,14], which allows to assess the stress effect on hydrogen diffusion in a ~10 μm scale. The neutron radiography results are then discussed in comparison with the finite element (FE) modeling. This coordinated research approach is expected to provide a better prediction of hydrogen diffusion behavior, which is the first step of the DHC failure.

A fully recrystallized Zircaloy-4 (1.41Sn-0.21Fe-0.11Cr-0.12O, wt%) sheet with a thickness of 4.5 mm was employed. The sheet was charged with pure hydrogen gas at 550 °C, followed by a homogenization heat treatment of 3 days at the same temperature in an Ar atmosphere. The hydrogen content was measured with six small specimens ($2 \times 2 \times 4.5\ mm^3$ in size) by hot gas extraction, presenting an average value of ~600 wppm. Then a notched sample was fabricated along the rolling direction, as shown in Fig. 1(a).

In order to evaluate the stress effect on hydrogen redistribution, it has been decided to examine the case of non-uniform stress field in the notch vicinity during a 3-point bending test. As shown in Fig. 1(b), the hydrogenated sample was heated up to 500 °C at a rate of 5 °C/min, followed by staying 1 h to dissolve hydrides and cooling down at a rate of 2 °C/min to 350 °C. This temperature is somewhat above reactor temperature; the hydrides re-precipitated, with ~230 wppm hydrogen remaining diffusible [15]. Then a load of 270 N was applied, with the temperature hold to let hydrogen in solid solution diffuses under the sole effect of external stress. After 10 h, the heating was shut down with the loading held until the temperature dropped down to 100 °C. The cooling process was planned in a way to conserve as best as possible the situation at the end of the temperature plateau, i.e. it was assumed to be fast enough so that no significant further hydrogen diffusion would take place after the cooling has started. Further, the load was held during cooling down to avoid any further re-distribution of the hydrogen concentration. In the holding phase at the temperature plateau, the stress-driven diffusing hydrogen arrived in the area of elevated stress and then precipitated there, since the solubility changes very little by applied stress [16]. The re-distributed hydrides by external stress were supposed to be retained by holding the stress applied during cooling. The bending test was conducted in a meso-scale thermo-mechanical equipment, which was constructed especially for in-situ neutron radiography and currently is still in the phase of ex-situ tests. Fig. 2 presents the local stress field in the notch vicinity, which was submitted to tensile stress with the first and third principal stress mainly on the notch head whilst the second principal stress on the shoulder.

The concentration field of hydrogen in the notch vicinity, before and after the test, was examined by neutron radiography using the

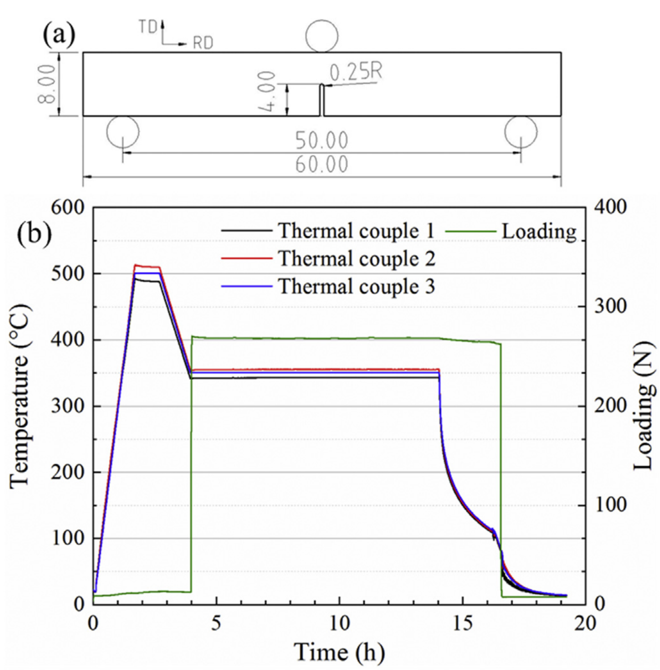


Fig. 1. (a) Geometry of the notched sample (in mm) and (b) data acquisition for the thermo-mechanical 3-point bending test.

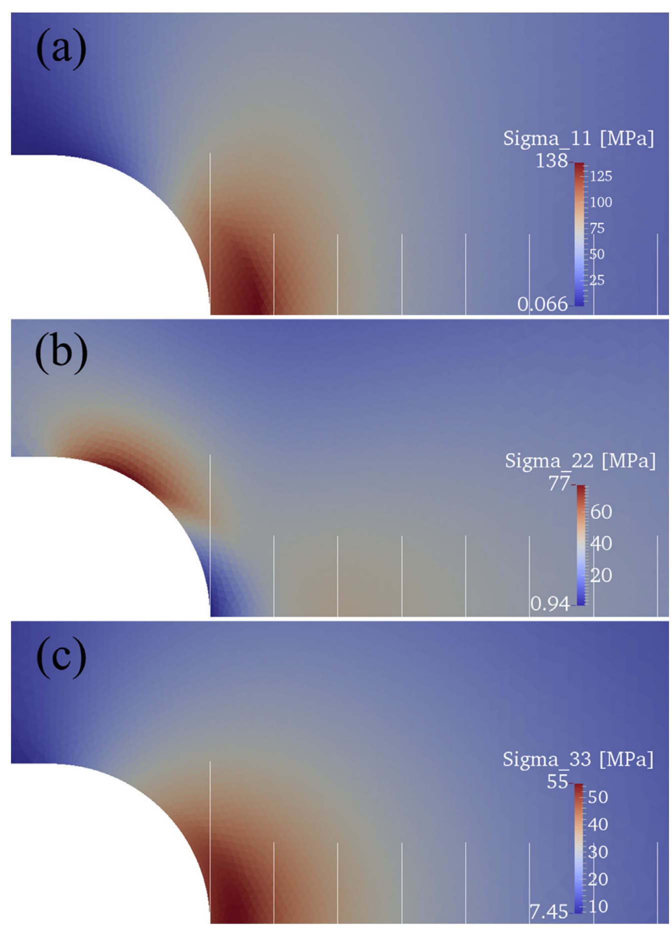


Fig. 2. Cropped stress fields of the notch vicinity from a half-scale FE computation (spatial scale bar spacing 0.1 mm) with σ_{11}, σ_{22} and σ_{33} along the rolling, transverse and normal direction, respectively.