Experimental report

13/09/2023

Proposal: 1-06-19 **Council:** 4/2023

Title: Investigation of the orientation dependence of hydrogen diffusion in nuclear fuel cladding tubes made ofzirconium alloys

Research area: Materials

This proposal is a new proposal

Main proposer: **Mirco GROSSE**

Experimental team: Mirco GROSSE
Sarah WEICK

Local contacts: Alessandro TENGATTINI

Samples: Zry-4 (Zr 1.5%Sn)

Instrument	Requested days	Allocated days	From	To
NEXT	3	2	30/08/2023	01/09/2023

Abstract:

The anisotropy of the hydrogen diffusion dynamics in hexagonal zirconium-based nuclear fuel cladding tube materials shall be investigated in-situ by means of neutron imaging. Due to their fabrication process they have a strong texture and thus the grain orientation in axial direction of the hexagonal lattice differs from the orientation in circumferential direction. As a consequence, the anisotropic hydrogen diffusion process differs between different cladding tube materials and between the direction of the uptake process. In order to model the hydrogen behaviour during long term dry storage of spent nuclear fuels, the anisotropy effect of the hydrogen diffusivity has to be determined precisely. Therefore, neutron imaging is the best and most precise measurement and visualisation method to record the process in-situ.

EXPERIMENTAL REPORT

Proposal number:	1-06-19		
Experiment title:	**Investigation of the orientation dependence of hydrogen diffusion in nuclear fuel cladding tubes made of zirconium alloys**		
Instrument	NEXT		
Dates of experiment:	30./08 – 01/09/2023	**Date of report:** **14/09/2023**	
Experimental team: **Names**	**Addresses**		
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The goal of the experiments was measuring the diffusion of hydrogen in nuclear fuel cladding tubes made of the zirconium alloy ZIRCALOY-4 (1.3% Sn, 0.3% Fe, 0.1% Cr, balance: Zr) in axial and circumferential direction. In order to fit the thermal expansion between the UO_2 pellets and the cladding tube material, the c-axis of the hexagonal zirconium lattice has to be oriented in the circumferential direction of the tube. Due to this strong texture, the diffusion coefficients in circumferential and in axial direction differs.
In order to measure the diffusion in circumferential direction, a 4.5 mm wide ring were cut from a commercial cladding tube. The ring sample was pre-oxidized to prevent hydrogen uptake. This ring was cut in axial direction too at one side. Here a metallic surface suitable for hydrogen uptake is produced. Into this cut a pre-hydrided wedge of zirconium was pressed. This zirconium wedge serves as hydrogen source. Fig. 1a) show a radiograph of this setup.
The sample was heated up to 425°C using a standard ILL vacuum furnace. Then, the sample was annealed at this temperature for 12.5 h.
Fig. 1 b) were obtained by referencing the neutron radiograph taken at the end of the test by the radiograph taken at the beginning. The contrast was calculated by:

$$C(x,y) = \frac{I(x,y)_{end\ of\ the\ test} - I(x,y)_{dark\ current}}{I(x,y)_{begin\ of\ the\ test} - I(x,y)_{dark\ current}}$$

C is the contrast, *x*,*y* the pixel coordinates and *I* the Intensity. Only the changes in the total macroscopic neutron cross sections during the annealing are visible. The pre-hydrided Zr wedge became brighter. It means, its total macroscopic neutron cross section is reduced by reduction of the hydrogen concentration. No changes in the ring shaped sample are visible. It shows that no hydrogen was picked up by the sample. The hydrogen released by the hydrogen source was not absorbed by the sample but was released into the furnace and pumped away.

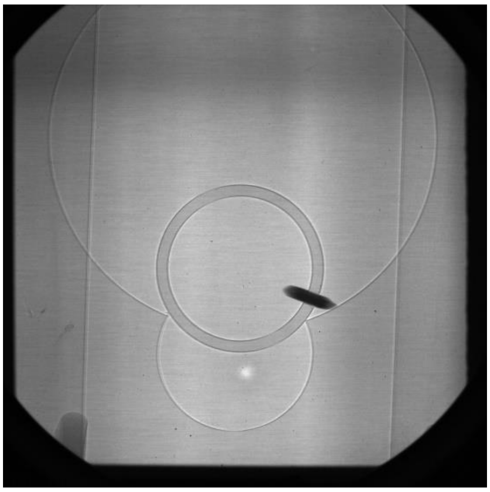

Fig. 1 a) Neutron radiograph of a cladding tube ring for investigation of the hydrogen diffusion in circumferential direction.

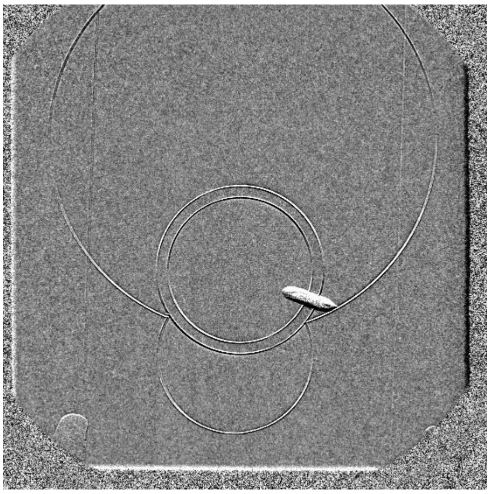

Fig. 1 b) Changes of the neutron radiograph between the beginning and the end of the test.

Another setup was tested using ZrH_2 powder as hydrogen source (see Fig. 1 a)). The setup allows hydrogen diffusion in axial direction. The oxide layer of the pre-oxidized sample was removed at the lower base plane standing in the powder bed by grinding. Here again, hydrogen is released by the hydrogen source but was not absorbed by the sample which becomes visible by the brighter grey scale of the referenced image. No changes in the grey scale of the sample is visible.

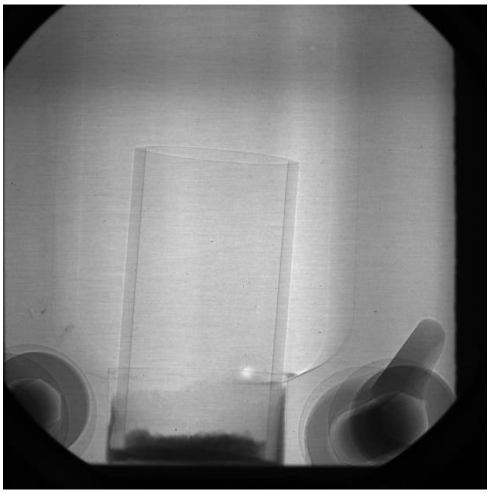

Fig. 2 a) Neutron radiograph of the cladding tube segment for investigation of the hydrogen diffusion in axial direction.

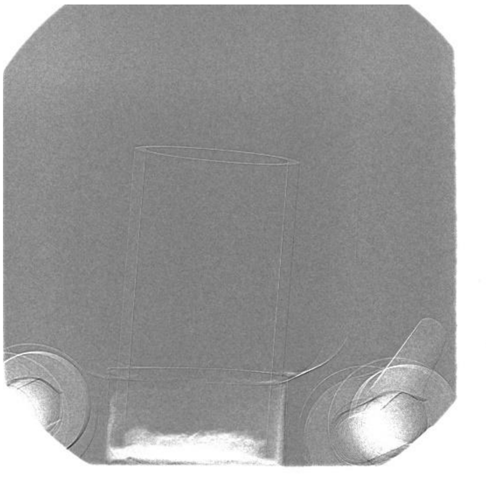

Fig. 2 b) Changes of the neutron radiograph between the beginning and the end of the test.

Obviously, the leak tightness of the sample holder was not good enough. According Sieverts' law $c_H^{metal} = \sqrt{k_s\, p_{H_2}}$, the hydrogen partial pressure p_{H_2} inside the sample holder equal to the total furnace pressure of several 10^{-5} mbar. Therefore, the equilibrium hydrogen concentration in the metallic zirconium alloy c_H^{metal} is much below the concentration detection limit of these measurements.

The experiments should be repeated with leak tight sample holders keeping the hydrogen partial pressure in the sample environment. Such sample holder is now under construction at KIT.