



Thermal expansion behavior of δ -zirconium hydrides: Comparison of δ hydride powder and platelets[☆]



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ARTICLE INFO

Article history:

Received 20 March 2018

Received in revised form

1 July 2018

Accepted 12 July 2018

Keywords:

Zirconium hydrides

Thermal expansion coefficient

Lattice parameter

δ hydrides

Synchrotron radiation

Mechanical strain

Dissolution and precipitation

ABSTRACT

The thermal expansion coefficient of δ hydrides and the evolution of the d-spacing of δ hydride platelets in Zircaloy-4 during heat treatments were investigated by conducting synchrotron x-ray diffraction experiments. Identical experiments enabled a direct comparison of the d-spacing of δ zirconium hydride platelets with the d-spacing of the powder δ hydrides. By analyzing the experimental data of this study and the data available in the literature, the thermal expansion coefficient of pure δ hydrides was determined as $14.1 \cdot 10^{-6} \text{ } ^\circ\text{C}^{-1}$. A direct comparison of the d-spacings of δ hydride platelets in CWSR Zry-4 sheet and the powder δ hydride samples showed an evolution of temperature-dependent strains within the δ hydride precipitates in which the strain components normal to the platelet edges exceed that normal to the platelet face during cooling/precipitation but *not* during heating/dissolution above approx. $200 \text{ } ^\circ\text{C}$.

Published by Elsevier B.V.

1. Introduction

The mechanical integrity of zirconium alloy nuclear fuel cladding may be compromised by the embrittling effects of δ zirconium hydrides at operation, storage, and transportation conditions [1–3]. While a light-water nuclear reactor (LWR) is operational, the zirconium alloy undergoes water-side corrosion which causes cladding oxidation and hydrogen release [4]. A portion of the released hydrogen diffuses into the cladding and precipitates as δ hydride phase, in the form of hydride platelets [5], hydride rim (a dense hydride platelet region) [6], and hydride blister (bulk hydride region) [7] if the hydrogen concentration in the cladding exceeds the solubility limit at corresponding temperatures. The presence of

these features may deteriorate the mechanical response of the cladding by crack initiation and loss-of-ductility during the nuclear fuel cycle [1,2,8], leading to potential nuclear fuel failures.

The δ -hydride (FCC) platelets and α zirconium (HCP) exhibit a closed-packed plane orientation relationship of $\delta\{111\}||\alpha\{002\}$ and have large anisotropic misfit strains associated with 17.2% volume difference between two phases [9]. The misfit strain is accommodated by the elastic and plastic deformation of δ hydride and α zirconium while δ hydrides are formed in α zirconium [10–12]. During their formation, the difference in the thermal expansion coefficients of δ hydride and α zirconium will also introduce strains due to the thermal-mismatch.

Synchrotron x-ray diffraction studies reveal that the evolution of the d-spacings of δ hydrides is complex during their dissolution and precipitation in α zirconium matrix [13–18]. Capturing such strain evolution requires usage of appropriate material properties of α zirconium and δ hydride. The α zirconium properties have already been well established in the literature [19], but the properties of δ hydrides involve a large uncertainty; in particular, values of the thermal expansion coefficient of δ hydrides range from 2.98 to $30 \cdot 10^{-6} \text{ } ^\circ\text{C}^{-1}$ [20–22]. A reliable value for the thermal expansion coefficient for δ hydrides is required for the accurate depiction of the mechanical strain evolution during dissolution and precipitation of δ hydrides.

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The thermal expansion coefficient of δ hydrides precipitates has been studied with the laboratory X-ray sources [20–23], but the energy resolution of conventional X-ray sources is too low to resolve the hydride peaks in detail if more than one hydride phase exists [24]. Synchrotron radiation source is capable of generating high-energy X-rays with high-brilliance which provides a more accurate data for an analysis of the lattice parameters of the dilute phases.

This manuscript investigated the thermal expansion behavior of bulk δ hydrides and δ hydride platelets precipitated in Zircaloy-4 matrix by performing high-energy synchrotron X-ray diffraction experiments. All samples were subject to identical thermal treatments while the diffraction patterns were acquired. First, the thermal expansion coefficient of δ hydrides was determined using bulk δ hydride powder samples. Second, evolution of the d-spacing of δ hydride precipitates in Zircaloy-4 was investigated by taking the bulk δ hydride results as reference. The results are discussed in terms of the previous thermal expansion data presented in the literature.

The outcome of this study extends our understanding of the behavior of hydride precipitates in zirconium alloys and support the computational studies of the mechanical integrity of the nuclear fuel cladding rods of LWRs due to the δ hydride formation and dissolution.

2. Method and procedures

2.1. Materials

This study utilized bulk hydride samples in powder form and a piece of a cold-worked stress-relieved (CWSR) Zircaloy-4 of flat sheet with a nominal thickness of 0.67 mm. Two types of samples were used in powder form: (i) δ hydride and Zircaloy-4 mixture (δ +Zry4) that contains 89 wt % of δ and less than 11 wt % of α zirconium phases and (ii) an ϵ hydride [25]. Zircaloy-4 discs (Zr–1.27Sn–0.22Fe–0.11Cr–0.118O) with a diameter of 12.7 mm and a thickness of 1 mm were employed to make powder samples after hydriding treatment.

Bulk hydride samples were fabricated as follows: (i) Zircaloy-4 samples were placed in a quartz boat and into a stainless steel vacuum chamber after removal of the native oxide layer by mechanical polishing; (ii) the chamber was evacuated down to 4 mTorr to produce an approximately oxygen-free environment, followed by several purges using ultra-high purity argon (99.999%); (iii) hydrogen with a purity of 99.9995% was introduced into the sealed chamber up to the pre-calculated pressure at room temperature; and (iv) the chamber was heated up to 400 °C at a rate of 10 °C/s and from 400 to 800 °C at a rate of 2.5 °C/min [26].

The hydrogen contents of samples were determined by using vacuum extraction method by Luvak Inc. (Boylston, MA). The average stoichiometry coefficients (r) were calculated as 1.61 ± 0.03 for δ +Zry4 and 1.79 ± 0.04 for ϵ hydride using hydrogen content of the samples, sample weight, and the nominal composition of zircaloy-4 [25].

After hydriding, the samples were characterized by using an electron back-scattering diffraction for the δ +Zry4 sample and the scanning electron microscopy (SEM) for the ϵ sample¹ (see in Fig. 1a and b). Both samples exhibited a polycrystalline structure after hydriding with an average equiaxed grain size of 5 μ m. The α zirconium grains were observed between δ hydride grains in δ +Zry4. The sample containing ϵ -hydrides showed the band structure of ϵ

hydrides, consistent with previously published results [21,27,28]. Bulk hydride samples were ground using mortar and pestle to prepare powder samples as shown in Fig. 1c². The powder sizes were ranged from 2.9 μ m to 16.35 μ m with average size \sim (5.20) $8.36 \pm 2.39 \mu$ m.

The sample from a pre-hydrated CWSR Zircaloy-4 flat sheet has a crystallographic texture similar to a typical CWSR Zircaloy-4 nuclear fuel cladding with Kearns' parameters of $f_N = 0.59$, $f_L = 0.05$, and $f_T = 0.31$ in the normal, longitudinal/rolling, and transverse directions, respectively [29]. The nominal dimensions of the sample were 1, 1, and 0.5 mm in length, height, and thickness, respectively. The sample was gas charged and contained 150 wt ppm of hydrogen. The details of the flat sheet sample and hydriding procedure was reported elsewhere [30]. Fig. 2 shows the resultant "in-plane"³ hydride microstructure after hydriding process where the platelet faces of the macroscopic hydrides were aligned parallel to the transverse direction (TD) and perpendicular to the normal direction (ND).

All samples were encapsulated in clear pyrex ampoules with an inner diameter of 1 mm, the wall thickness of 0.2 mm, and the length of 100 mm to eliminate the oxidation of the powder samples during X-ray diffraction experiments at elevated temperatures. Ampoules containing samples were subjected to three cycles involving a vacuum at 1.3 Pa and a subsequent flushed with Ar. After flushing with argon gas, ampoules were evacuated down to 33×10^{-3} Pa and sealed. During experiments, no detectable oxidation of samples was observed in the diffraction patterns.

2.2. Synchrotron X-ray diffraction experiments

Synchrotron radiation diffraction experiments were conducted at the beamline 6-ID-D at Advanced Photon Source at Argonne National Laboratory. Schematics of the experimental setup of the powder samples and the orientation of the incoming X-rays are shown in Fig. 3. A $200 \times 200 \mu\text{m}^2$ rectangular 100.379 keV X-ray beam transmitted through the ampoules and the powder samples generating Debye-Scherrer diffraction rings of the powder samples on an area detector. To increase the statistics of diffraction data acquisition, the diffraction patterns were acquired by 40 frames with 1 s exposure for each frame and were added up to generate one diffraction pattern.

Fig. 4a shows the schematics of the X-ray diffraction experiment of the sample from the Zircaloy-4 flat sheet. That sample was also placed into the ampoule where the incident X-ray was perpendicular to the transverse-normal plane of the sheet sample as shown in Fig. 4b. This orientation ensured to acquire the diffraction signals from the hydride platelets' face and edges to the area detector. In so doing, the d-spacing evolution of particular crystallographic planes of hydride platelets' face and edges were acquired during heat treatment.

The ampoules containing samples were located inside a furnace which provided the heat treatment shown in Fig. 5a to capture the change in the d-spacings. During the heating stage, samples were heated with a rate of 10 °C/min to a maximum temperature of 500 °C for δ +Zry4 powder and 600 °C for ϵ hydride powder. At each 50 °C interval, the samples were soaked for 5 min for thermal equilibrium and a diffraction pattern was acquired. The samples were subsequently cooled to 100 °C with a rate of 1 °C/min and the

¹ For ϵ hydride sample, roughness of the specimen surface prevented an accurate EBSD analysis, please see the details in Ref. [25].

³ In-plane hydrides corresponds to the circumferential hydrides in the tube geometry of nuclear fuel cladding. In-plane (or circumferential) hydride microstructure is caused by the crystallographic texture of the samples.

² In this study, we did not quantify that the powder preparation process may cause changes in the stoichiometry of the samples.

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