



In situ micropillar deformation of hydrides in Zircaloy-4

H.E. Weekes,^a V.A. Vorontsov,^a I.P. Dolbnya,^b J.D. Plummer,^a F. Giuliani,^a T.B. Britton^a and D. Dye^{a,*}

^aDepartment of Materials, Royal School of Mines, Imperial College London, Prince Consort Road, London SW7 2BP, UK

^bDiamond Light Source, Harwell Science and Innovation Campus, Didcot OX11 0DE, UK

Received 1 December 2014; revised 6 March 2015; accepted 23 March 2015

Available online 13 April 2015

Abstract—Deformation of hydrided Zircaloy-4 has been examined using *in situ* loading of hydrided micropillars in the scanning electron microscope and using synchrotron X-ray Laue microbeam diffraction. Results suggest that both the matrix and hydride can co-deform, with storage of deformation defects observed within the hydrides, which were twinned. Hydrides placed at the plane of maximum shear stress showed deformation within the hydride packet, whilst packets in other pillars arrested the propagation of shear bands. X-ray Laue peak broadening, prior to deformation, was associated with the precipitation of hydrides, and during deformation plastic rotation and broadening of both the matrix and hydride peaks were observed. Post-mortem TEM of the deformed pillars has indicated a greater density of dislocations associated with the precipitated hydride packets, while the observed broadening of the hydride electron diffraction spots further suggests that plastic strain gradients were induced in the hydrides by compression.

© 2015 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

Keywords: Zirconium; Synchrotron diffraction; TEM; Micromechanics; Hydrides

1. Introduction

Zirconium alloys are widely used as nuclear fuel cladding material and readily absorb hydrogen during service in a pressurised water reactor (PWR) primary circuit environment. Possible mechanisms allowing hydrogen to be either generated in or introduced into the clad material include corrosion, radiolysis of water, fuel oxidation and dissolved coolant hydrogen [1]. Although able to accept up to 50 at.% hydrogen in solid solution at temperatures above 500 °C, as temperature decreases, so does the alloy's ability to accommodate hydrogen in solution. At room temperature, the solubility may be as low as 10^{−4} at.% [2]. This leads to the formation of embrittling zirconium hydrides, particularly during cooling from the operating temperature (~350 °C).

Hydride formation is generally considered detrimental to mechanical properties and results in decreases in fracture strength, impact strength and tensile ductility at slow strain rates [2]. The extent of hydride embrittlement on the accommodating Zr matrix is found to be critically dependent on both their morphology and orientation relative to the applied load direction [2]. Although the texture of zirconium alloys can be controlled so as to promote hydride precipitation in relatively benign orientations, hydrides can reorient to undesirable orientations (perpendicular to load direction) during cooling under an applied tensile stress

[3–5]. Enhanced hydride nucleation is readily observed within the highly stressed region of a crack tip and, together with hydride reorientation under a tensile stress, results in the initiation of the intermittent crack growth process known as delayed hydride cracking (DHC). DHC was initially not considered a major issue at temperatures below 200 °C [6], but reports of component failures after long term room temperature storage [7] have led to greater attention to this phenomenon.

Individual zirconium hydrides display a characteristic acicular, or plate-like, morphology with a habit plane parallel to {0002}_{Zr}. A {10 $\bar{1}$ 7}_{Zr} habit plane is found for the macroscopic hydride packets observed by optical microscopy, which was studied by transmission electron microscopy (TEM) by Chung et al. [8], Fig. 1(a). The plate size and morphology are a consequence of transformation strains, and have been rationalised with the phenomenological theory of martensite transformations [9–12]. Individual nano-hydride platelets are autocatalytically nucleated in macroscopic hydride packets, several μm in length, *via* a process of strain accommodation. The sharp tips of these nano-hydrides have been observed to punch dislocation loops into the surrounding Zr matrix [13,14], illustrated in Fig. 1(b).¹ These shear loops should be

¹The reader should note that there is a concern in the community as to whether the hydrides studied in Carpenter et al. [14] were artefacts introduced by polishing. In addition, Carpenter et al. studied γ hydrides, not δ-ZrH.

* Corresponding author; e-mail: david.dye@imperial.ac.uk

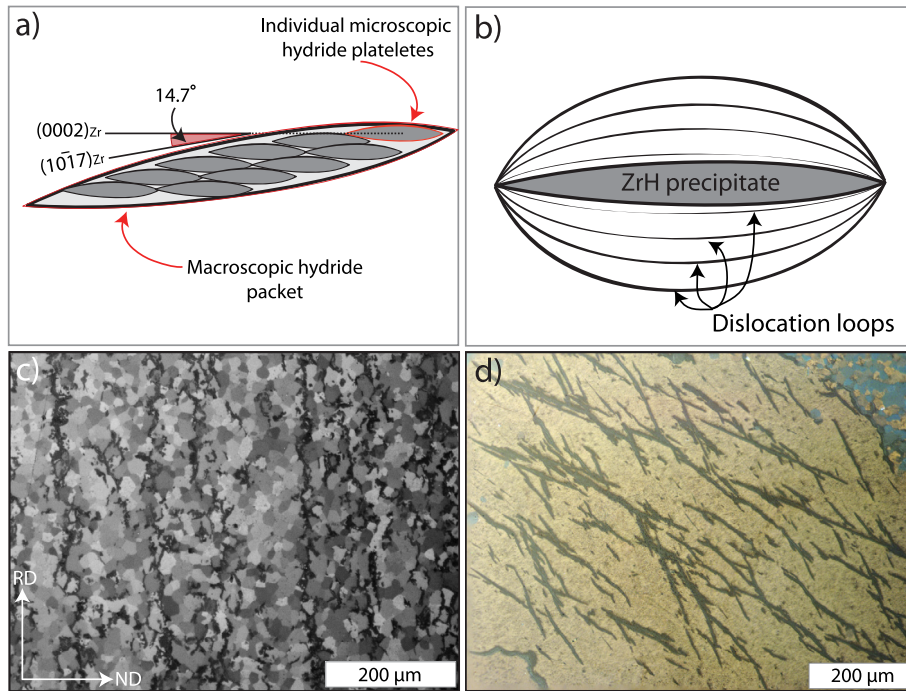


Fig. 1. (a) Hydride habit planes in α -Zr: $\{10\bar{1}7\}$ for macroscopic hydride packets and $\{0002\}$ for microscopic hydride platelets (illustration adapted from [8]), (b) dislocations generated by the nucleation of hydride precipitates in the Zr matrix (illustration adapted from [14]), (c) and (d) optical micrographs of electrolytically hydrided, hot-rolled and recrystallised Zircaloy-4 before (c) and after (d) annealing heat treatment to promote both grain growth and intragranular hydride precipitation.

distinguished from the prismatic loops that can be observed due to irradiation. It should also be noted that the dislocations observed around surface hydrides generated by electropolishing or around hydrides induced by quenching may not be representative of the state where hydrides form on slow cooling.

In recent years, the field of experimental small scale micromechanics has grown dramatically. Demonstration experiments performed on the deformation of pillars, 1–10 μm in width, have highlighted the potential of *in situ* synchrotron X-ray Laue microbeams to probe the deformation response of materials [15–17], although only ideal or ‘model’ materials have been examined. Changes in lattice structures, such as rotation gradients and sub-grain formation, have been observed through diffraction peak evolution. The sensitivity of microbeam diffraction to defect structures has also enabled the study of dislocation density evolution through both the streaking and broadening of diffraction peaks. It is therefore timely to ask if such experiments can provide useful insights into the deformation response of real, industrial, micromechanics problems such as that of hydrides in zirconium.

In the present work, we examine the deformation behaviour of electrolytically hydrided Zircaloy-4 Focussed Ion Beam (FIB) milled micropillars. Loaded in compression and oriented at approximately 45° to the loading axis, the hydride plates were positioned near the plane of maximum shear. First, *in situ* testing within a Scanning Electron Microscope (SEM) chamber is presented in order to elucidate the general deformation behaviour of the pillars. Then, the evolution of the diffraction patterns obtained by *in situ* microbeam synchrotron Laue diffraction is examined. Finally, the post-loading dislocation states are explored via

(Scanning) Transmission Electron Microscopy, (S) TEM, using thin foils FIB-milled from the deformed pillars.

2. Experimental

Electrolytic hydriding was carried out in dilute sulphuric acid (1% H_2SO_4) at 65°C ($\pm 5^\circ\text{C}$) using a current density of 2 kAm^{-2} . Heat treatment then interdiffused the hydrogen uniformly through the sample and a slow furnace cool promoted the formation of stable δ hydrides. The Zr texture acquired through hot rolling aligns the basal poles perpendicular to the rolling direction ($\pm 20^\circ$) [18] and Fig. 1(c) highlights the resulting hydride alignment due to the known $(0002)_{\text{Zr}} \parallel (111)_{\text{ZrH}}$ orientation relationship. Hydride precipitation readily occurs at α -Zr grain boundaries, and to avoid this the sample was annealed at 830°C for 24 h to promote both grain growth and precipitation of intragranular hydrides. Fig. 1(d) shows the microstructure and hydrides after the applied heat treatment.

A grain of interest with its $\{0002\}$ close to 45° from the sample surface was identified using electron backscatter diffraction (EBSD) and, for pillars to be used during *in situ* diffraction experiments, sliced to produce a thin wedge with a $50\text{ }\mu\text{m}$ platform edge for fabrication. *In situ* SEM pillars were produced directly from the bulk. Slice and view FIB milling, Fig. 2(a), coupled with sample surface EBSD analysis located hydrides oriented near 45° from the loading axis. Perpendicular focussed ion beam (FIB) milling (30 keV with a final polishing step of 30 pA), was then used to produce pillars with a slight taper and aspect ratio of $\sim 3:1$. In this geometry, one end of the pillar remained part of the bulk while the other was a free surface.

Download English Version:

<https://daneshyari.com/en/article/1445388>

Download Persian Version:

<https://daneshyari.com/article/1445388>

[Daneshyari.com](https://daneshyari.com)