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Study on the morphology of bulk hydrides by synchrotron X-ray tomography

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ABSTRACT

The morphologies of hydrides was studied using synchrotron X-ray at the European Synchrotron Radiation Facility (ESRF) using Diffraction Contrast Tomography (DCT). Diffraction Contrast Tomography is a relatively new technique that uses simultaneous collection of tomographic and diffraction data to reconstruct the 3D microstructural volume; in this case precipitates and matrix grains, while providing crystallographic orientation of neighbouring matrix grains. Previously the method was limited to light metals such as Al or Mg (due to the use of 8–30 keV X-rays). Here we show the first DCT measurements in a high atomic number metal zirconium using high energy X-rays (60 keV).

1. Introduction

Zirconium alloys are commonly used in nuclear reactors due to a combination of good mechanical properties and low neutron absorption cross section. The absorption of hydrogen during service in water at elevated temperatures leads to the precipitation of a brittle hydride phase in zirconium component upon reactor shutdown [1]. The orientation and distribution of the hydride phase are directly related to the mechanical properties of the material. Tests on fuel claddings showed a drastic decrease in tensile strength when hydrides precipitated in the radial direction (i.e., such that the hydride platelet normal is parallel to the applied stress during the test) [2]. Therefore, the morphology of the precipitated hydrides in zirconium are of considerable interest.

The morphology of hydrides includes two aspects at different length scales. First, the morphology of a single hydride is a needle shaped single crystal particle with a crystallographic orientation relationship with the matrix zirconium grain [3]; second, these acicular hydride crystals often form in colonies [3] and collectively they have an orientation and distribution.

The crystallographic orientation between hydrides and neighbouring zirconium grains is only available using techniques such as Transmission Electron Microscope (TEM) and Electron Backscatter Diffraction (EBSD). In TEM the relationship was first determined to be (0001)-Zr ‖ {111}-ZrH1.5 [3]. With EBSD it was found that the habit plane of hydride was related to the location where it precipitated: intragranular hydrides was confirmed to have the (0001)-Zr ‖ {111}-ZrH1.5 relationship, while intergranular hydrides have other orientation relationships except that one [4,5,6].

The collective orientation distributions of hydrides are often studied by 2D metallographic examinations [7], which is both laborious and does not provide the crystallographic orientations of the grains. EBSD is a big improvement in both aspects, but the 2D cross section of a hydride still does not readily reveal the 3D orientation. Diffraction Contrast Tomography (DCT) is a relatively new technique that uses simultaneous collection of tomographic and diffraction data. The technique uses a parallel monochromatic X-ray beam. The sample is mounted on a rotating stage, and a high resolution 2D detector is placed close to the sample. During rotation of the sample, diffraction occurs on grains satisfying the Bragg condition. The shape of the diffracted spot is close to the shape of the grain viewed from the diffracted beam direction. The clear benefits of DCT versus other techniques are the 3D information it provides and the short time it needs for a reconstruction. The only possible alternative to such 3D X-ray techniques is depth profiling of the same volume size in a Scanning Electron Microscope (SEM) using ion beams [8], which would take much longer and is necessarily destructive of the sample.

2. Material and Methods

2.1. Sample Preparation

The material for this study was a Zircaloy-2 slab with a measured composition Zr, 1.43–1.45 wt% Sn, 0.13–0.14 wt% Fe,0.10 wt% Cr, 0.05 wt% Ni, 1260–1440 wppm O and 150–160 wppm C. The sample were electrolytically charged in a 0.2 M solution of H_2SO_4 at 80 ± 5 °C for 48 h followed by solution treating at 334 °C for 40 h to give a

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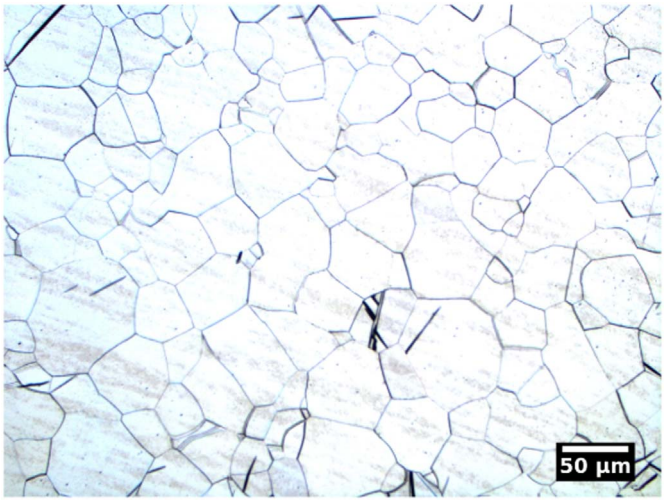

Fig. 1. α Zirconium grains under optical microscope.

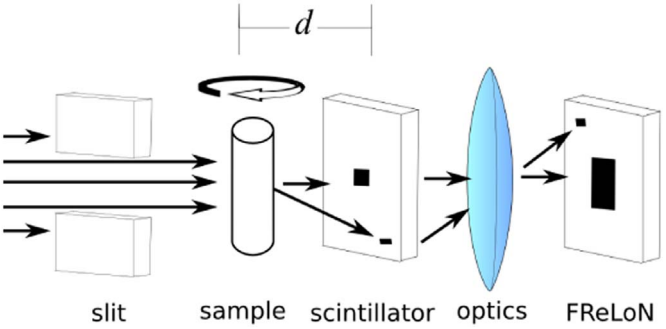

Fig. 2. Schematic illustration of the diffraction setup.

Fig. 3. Computed cross section of absorption contrast showing dark hydride phase.

nominal hydrogen concentration of 100 wppm [9]. The material has an equiaxed grains as shown in Fig. 1. A 20 mm long stick with a cross section of 450 μm by 450 μm was cut with a fine saw blade and mounted vertically for the diffraction tests.

2.2. Experimental Setup

The experiment was carried out at the Id11 beam-line in ESRF. The diffraction setup is illustrated in Fig. 2. The beam is a parallel monochromatic 60 keV X-ray with a size of 800 μm by 500 μm. The sample volume exposed in the beam is thus around 0.1 mm^3. The sample to scintillator distance d was 10 mm to allow capturing of the diffraction spots and was increased to 500 mm for conventional phase contrast tomography (PCT). The PCT was used to determine the location of the hydride phase, which did not provide sufficient diffraction signal for accurate DCT. The scintillator screen used to convert X-rays into visible light consists of a gadolinium oxysulphide layer of 60 μm thickness. The image on the scintillator is recorded with a Fast Readout Low Noise detector of the ESRF (FReLoN), which has a 14 bit dynamic CCD camera, with a 2048 by 2048 pixel chip [10]. The setup gave an effective pixel size of 1.4 μm for the DCT measurements and 0.7 μm for the PCT measurements.

2.3. Data Processing

2.3.1. Reconstruction of Hydrides

The Pagnin phase retrieval technique [11] was used in the phase contrast tomographical reconstruction of hydride. The volume expansion during hydride precipitation was calculated to be 17% [12], which allows the hydride to be clearly distinguished by the absorption difference between hydride and the zirconium matrix as shown in Fig. 3. The threshold for hydride phase was found by setting hydride voxel percentage equal to the known average hydride volume percentage. For 100 wppm weight percent hydrogen, the corresponding hydride volume percentage is 0.7%.

2.3.2. Reconstruction of Zirconium Grains

The shape of the zirconium grains and their positions and orientations were obtained with the DCT reconstruction, using an arithmetic reconstruction algorithm from all the diffracted spots. The reconstruction was done with the in-house DCT code developed at ESRF, with extra precision in crystal orientation achieved by taking advantage of the Friedel pairs [13]. The procedure was described in [14].

2.3.3. Misorientation Calculation

The zirconium grain boundaries was believed to play a significant role in the oxidation and deuterium ingress resistance of zirconium alloys [15]. Grain boundaries were characterised by the misorientation of the grains, which was best described by the Rodrigues vector. The smallest Rodrigues vector was calculated following the procedure by Rollett [16], which was summarized as follows:

(1). The orientation matrices for all symmetry operations were calculated according to Eq. (1), where O_c is the symmetry operator and g is the grain orientation matrix, Δg is the relative orientation matrix, subscript i and j are the grain index, A and B denote the two coordinate systems. The symmetry operator is from [17].
(2). For each orientation matrix, a corresponding Rodrigues vector was calculated, the smallest Rodrigues vector and the corresponding symmetry operator were recorded.

$$\Delta g = (O_{ci} g_B)(O_{cj} g_A)^{-1} \quad (1)$$

2.4. Collapsed Hough Transform

The Hough technique is well known for edge detection [18] and is particularly useful for computing a global description of a linear or planar feature given (possibly noisy) measurements. For the purpose of studying the local orientation of hydride pixels, the Hough transform was modified as the following Collapsed Hough Transform procedure:

(1). Discretise the orientation space into finite number of orientations.
(2). Move the origin to the hydride pixel of interest (the red dot shown in Fig. 4), perform a Hough transform on pixels within a distance of