



# Hydrogen evolution and its effects on cold rolling behavior in commercial pure titanium

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## ABSTRACT

The microstructural evolution induced by hydrogen in the commercially pure Ti50A and its effect on the deformation mechanisms during symmetrical cold rolling have been investigated by means of X-ray diffraction, scanning electron microscopy equipped with electron backscatter diffraction and transmission electronic microscopy. Upon charging by electrolytic method, both  $\delta$ -TiH<sub>x</sub> and  $\epsilon$ -TiH<sub>2</sub> titanium hydrides were found to precipitate in the grain boundaries as well as within the  $\alpha$ -phase matrix. In addition to the refinement of the microstructure, microstructural analyses of the cold rolled samples highlighted the formation of  $\{10\bar{1}2\}\langle\bar{1}011\rangle$  tension twins and  $\{11\bar{2}2\}\langle11\bar{2}3\rangle$  compression twins in the hydrogenated samples rather than the  $\{11\bar{2}1\}\langle\bar{1}\bar{1}26\rangle$  tension twins usually activated in the hydrogen-free Ti50A. No evidence of correlation between the hydrides and twin formations could be detected however measurements of the local lattice curvature and strain gradients from EBSD analyses revealed a larger density of the geometrically necessary dislocations in the hydrogenated samples that could be attributed to the presence of hydrides and hydrogen dissolved in the solid solution  $\alpha$ -phase.

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## 1. Introduction

Titanium and titanium alloys have a high affinity for hydrogen and many studies have already successfully exploited hydrogen as a temporary element to process titanium components and to modify their properties [1–3]. Hydrogen is known to destabilize the hexagonal  $\alpha$ -phase and stabilize the softer body-centered cubic (bcc)  $\beta$ -phase in titanium alloys so that its addition increases the proportion of the  $\beta$ -phase. Therefore, earlier studies have demonstrated that the addition of a certain amount of hydrogen enhances the ductility and improves the workability of  $\alpha$  and  $\alpha + \beta$  titanium alloys [4–6]. The commercially pure titanium Ti50A (*ASTM Grade 2*) has been widely used in key engineering applications covering a variety of areas, such as chemical industrial, marine, biomaterials, etc., due to its unique combination of mechanical and chemical properties [7,8]. In most of these applications, the components operate in environment that can act as sources of hydrogen after dissociation of the H<sub>2</sub> molecules on Ti surfaces [9]. Consequently, understanding the interaction of hydrogen with this material is important so that its properties can be controlled and reliably predicted.

Recent developments in the study of hydrogen effects on dislocation nucleation using in situ electrochemical nanoindentation in various alloys have shown that hydrogen reduces the required stress for the onset of plasticity [10–12], which has been explained owing to a

consequence of a reduction in the shear modulus, dislocation line energy and stacking fault energy. Based on the concept of defactant (defects acting agents) [10,11], hydrogen was observed to facilitate the formation of vacancies, dislocations and stacking faults under plastic deformation. For example, Krystian et al. [13] have reported that the formation of superabundant vacancies can be produced under high-pressure torsion in the presence of hydrogen. Also, Deutges and coworkers [14], who studied the effect of hydrogen on the density and arrangement of dislocations in cold-rolled palladium-hydrogen alloys, have reported that hydrogen can increase the dislocation density and lead to homogeneously distributed dislocations throughout the whole material. However, few studies have focused on the effect of hydrogen on the cold deformation behavior in titanium-based materials. Therefore, the aim of the present work is to inspect how hydrogen acts on the cold rolling behaviors of commercial pure titanium Ti50A.

In previous studies, the hydrogen effects on the phase transformation, microstructure evolution and hydride formation mechanism in commercially pure titanium Ti40 [15,16] were investigated by means of X-ray diffraction (XRD) and transmission electron microscopy (TEM). The Scanning Electron Microscopy (SEM)-based Electron Backscatter Diffraction (EBSD) method is well placed to bridge between XRD and TEM methods by revealing spatial patterning in deformation structures over regions that span over several contiguous grains [17]. It is a powerful technique to automatically and quantitatively investigate the phase transformation, grain/subgrain size, point-to-point orientations, strain, and local texture [18]. The objective of the present investigation is to describe thoroughly the evolution of the

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microstructure of Ti50A induced by hydrogen and its effects on the cold rolling behavior using combined XRD, TEM and SEM-EBSD analyses.

## 2. Material and Experimental Details

### 2.1. Material and Processing

The material used in this study was the commercially pure titanium Ti50A in the form of 1.60 mm thick sheet supplied by TIMET in the annealed state. To emphasize the hydrogen-affected region, the thickness of the as-received (AR) samples mechanically cut from the sheet was reduced by mechanical polishing followed by an annealing treatment performed in the vacuum for 2 h at 350 °C in order to relieve the potential residual stresses generated during sample preparation. The sample surfaces were then polished using successively SiC papers of 300, 500, 1000, 2000 and 4000 grit to remove the oxidized and contaminated layers. Moreover, to minimize the undesired factors affecting hydrogen evolution in the material microstructure, the samples were vibration polished for 2 h to remove residual stresses and deformed layers. The final thickness of the sample was around 1.0 mm.

Hydrogen was introduced into the specimens by an electrolytic method. A platinum anode of cylindrical shape was used, and specimen was located in the center of the electrolytic cell as described in ref. [19]. Specimens ( $50 \times 15 \times 1.0 \text{ mm}^3$ ) were charged with hydrogen in an electrolyte consisting of 1/3 phosphoric acid (85%) and 2/3 glycerin (85%) [20] under an applied current density of 2 mA/cm<sup>2</sup> for either 72 h, 108 h or 168 h. The amount of hydrogen dissolved in the sample was measured via the inert gas fusion instrument (LECO TCH-600) at the University of Utah in Salt Lake City, U.S.A. To inspect the effect of hydrogen on the microstructure evolution in Ti50A upon cold deformation, the samples following hydrogenation were immediately deformed at room temperature by normal symmetric rolling using a fixed rolling speed of 50 mm/s. Thickness reduction ratio of 10 and 50% were carried out by unidirectional multi-pass rolling using ~5% reduction per pass.

### 2.2. Experimental Characterization

To examine the hydrogen evolution in Ti50A, XRD, SEM-EBSD and TEM analyses were performed immediately after hydrogen charging. The XRD measurements were undertaken using Cu-K<sub>α</sub> radiation with a wavelength of 0.154 nm. In order to index qualitatively EBSD patterns (Kikuchi patterns), the samples were electropolished at 5 °C and 30 V for 10 s in a solution of 100 ml perchloric acid and 900 ml methanol. The microstructures developed through hydrogen charging and cold rolling were examined using a JEOL-6500F field emission gun scanning electron microscope equipped with an EBSD acquisition camera and the AZtec acquisition software package (Oxford Instruments). Based on the grain size, beam control mode EBSD was used with a step size of 0.15 μm to minimize the measurement time and obtain relatively reliable results. For individual orientation of the locally deformed microstructure, the EBSD orientation data were analyzed using the Analysis Tools for Orientation Map (ATOM) software [21].

For TEM observations, thin foils were extracted from the hydrogenated samples by ionic micromachining from selected areas previously localized by SEM-EBSD. Extraction was performed with a Focused Ion Beam (FIB) Zeiss Auriga 40 equipped with both electronic and ionic columns (Gemini and Orsay Physics Cobra, respectively). While most of the machining process was undertaken with an ionic Ga beam of 30 kV, thin foils were finished under a beam of 2 kV before extraction to eliminate the amorphous layer that could form under the high-energy ion beam. Then, microstructures were observed in a Philips TEM 200 CX for both imaging and phase determination upon a working voltage set at 200 kV.

The Vickers hardness was measured on the specimen cross section using a Microhardness Tester (DIN50190/1) under a load of 50 g applied

during 10 s. The microhardness profile from the surface to a center of samples was performed in a zigzag mode at intervals of 0.05 mm.

## 3. Results and Discussion

### 3.1. Examination of the Hydrogen Evolution Using XRD, EBSD and TEM Characterizations

In electrolytic method, the influx of hydrogen into the sample consists of the following simple steps: 1) deposition of hydrogen ions onto the electrode surface; 2) consumption of one electron by each hydrogen ion to form atomic hydrogen; 3) adsorption of atomic hydrogen on the surface of sample electrode; 4) dissolution of hydrogen in the sample electrode surface; 5) diffusion of atomic hydrogen into the sample through the lattice and defects. In pure titanium and for low concentration, hydrogen atoms dissolved in the α-solid solution occupying interstitial sites of the hexagonal lattice and can be defined as quasi-mobile hydrogen atoms. For higher hydrogen concentration, above the expected terminal solubility limit, the formation of hydride takes place; these atoms bonded to Ti will be referred as bonded hydrogen. Values of the hydrogen concentration achieved in the current study are presented in Table 1. It is worth noting that a small amount of hydrogen was already detected in the as-received state. Moreover the dissolved hydrogen concentration increased linearly with the charging time, which indicates that lattice diffusion appears to be the rate controlling mechanism in the process of hydrogen charging in Ti50A.

The X-ray diffraction patterns after 72 h hydrogen charging time, as shown in Fig. 1, evidenced the precipitation of two types of hydrides, i.e., δ-TiH<sub>x</sub> and ε-TiH<sub>2</sub>, for a concentration of about 60 ppm. Using mechanical polishing, the thickness of the hydride layer as determined by XRD was estimated to be about 20 μm. The TiH<sub>x</sub> ( $1.5 \leq x \leq 1.99$ ) δ-hydride has an fcc lattice ( $a = 0.4445 \text{ nm}$ ) with the hydrogen atoms occupying the tetrahedral interstitial sites (CaF<sub>2</sub> structure), while the TiH<sub>2</sub> ε-hydride has a bct (body centered tetragonal) lattice with  $a = 0.312 \text{ nm}$  and  $c = 0.418 \text{ nm}$  (JCPDS-PDF 9-371) [22,23]. As observed in the XRD patterns, the intensity of the diffraction peaks corresponding to the hydride phases increased with the charging time, suggesting an increase in the volume fraction of hydrides formed in the sample. According to the binary constitution phase diagram of Ti–H system, the terminal hydrogen solubility in the alpha phase is only approximately 1500 ppm and decreases rapidly with decreasing temperature. At room temperature, the terminal hydrogen solubility in α-titanium is quite negligible (~8.35 ppm) [24]. Therefore in addition to hydrides formed a thin layer beneath the surface, the results described above suggest the existence of a supersaturated solution of hydrogen in titanium underneath this layer.

In addition, it can be noticed that the XRD peaks corresponding to the matrix α phase became broadened as the charging time increased. Williams et al. [25] have pointed that the α-titanium to the δ-TiH<sub>x</sub> phase transformation resulted in an approximately 17.2% volume expansion, and this volume effects associated with the hydride formation led to a local plastic deformation of the matrix accompanied by an increase in the dislocation density. Therefore, the broadening peaks of α-phase can be attributed to the occurrence of local deformation caused by the phase transformation within the sample.

To quantitatively investigate the hydrogen evolution, the microstructures of Ti50A samples after 72, 108 and 168 h hydrogen charging were characterized by EBSD technique. Due to necessary sample preparation for the EBSD measurements, a layer of approximately 10 μm thick

**Table 1**  
Hydrogen contents for different charging times.

Charging time	0 h	72 h	108 h	168 h
Hydrogen content, ppm	23.32 ± 2.30	65.17 ± 3.50	88.53 ± 4.00	138.05 ± 6.50

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