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Effect of interstitial oxygen and iron on deformation of Zr–2.5 wt% Nb

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ABSTRACT

A series of *in situ* compression tests have been carried out at room temperature on three dual-phase Zr–2.5 wt% Nb alloys with different concentrations of the interstitial alloying elements oxygen and iron. Oxygen and iron are potent strengthening additives in two-phase alloys with a hexagonal-close packed α phase and body-centred cubic β phase, such as the Zr–Nb and Ti–V–Al systems. The evolution of interphase stress, along with the progression of twinning, were monitored during deformation by neutron diffraction. The twin volume fraction measured following deformation was not strongly dependent on ingot chemistry, contrary to previously reported results. Annealing in the $\alpha+\beta$ and the β region allowed for variation in the initial residual stress distribution and crystallographic texture, respectively, and the effect of these parameters on deformation mode selection was also investigated. Interstitial oxygen has a greater strengthening effect on prismatic $\langle a\rangle$ slip than on basal $\langle a\rangle$ slip at room temperature. High impurity contents were not observed to suppress $\{10\bar{1}2\}\langle 10\bar{1}1\rangle$ tensile twinning. Despite being an α-stabilizer, oxygen is observed to strengthen the β phase under suitable heat treatment conditions.

1. Introduction

Zirconium and its alloys are widely used as structural materials in the nuclear industry. One such alloy, Zr–2.5 wt% Nb, is used primarily in pressure tubes in the CANDU reactor designs. The in-service properties of Zr–2.5 wt% Nb are impacted by the texture, microstructure, and intragranular stresses developed during fabrication [1]. At room temperature and at operating conditions, this alloy has a two phase microstructure, with the Nb-rich β phase comprising roughly 10 vol%. Alloying components include iron (<1300 wppm) and oxygen (between 900 and 1500 wppm) in the CANDU design [1]. Interstitial oxygen is known to increase the yield strength of α-zirconium (the Nb-poor phase), while iron is segregated to and strengthens the β-phase (identical to the case in Ti alloys [2]).

Studies on $(\alpha+\beta)$ Ti alloys have found that the presence of the β phase has a strong effect on the mechanical response of the α phase. The β phase has been reported to decrease the occurrence of both $\langle c+a\rangle$ slip and twinning in the α phase [3], while also increasing the activity of prism $\langle a\rangle$ slip [4]. Other studies have found that the presence of the β phase encourages twinning in the α phase [5,6]. The β phase is reported as either softer (in Zr [7] and Ti [3]), or harder (in Zr [8] and Ti [5]) than the α phase, depending on the specific alloy and sample history.

The role of interstitial oxygen has been previously investigated in titanium. A yield asymmetry has been noted in commercially pure titanium by Brandes et al. [9]. The origin of this asymmetry was attributed to the non-planar dislocation core of prism $\langle a\rangle$ dislocations in Ti, and the stabilizing effect of oxygen on the core structure of these dislocations [10]. Single crystal studies have shown that prism $\langle a\rangle$ slip is sensitive to oxygen content in Ti [2]. At increased temperatures, this effect is reduced and only a weak dependence is observed above 600 K. Oxygen has been observed to prevent cross-slip of dislocations in titanium, even at relatively high temperatures when the concentration is at least 1750 wppm, as indicated by a transition from wavy to straight flow of screw dislocations [10]. This suppression of cross-slip is also likely to impact Zr alloys, which also exhibit wavy slip traces following basal slip [11].

The development of deformation texture in hcp materials is dependent on the balance between slip and twinning deformation modes. The activation of one mode over another depends on their relative critical resolved shear stresses (CRSS), as well as the constraints of surrounding grains (so-called Type II stress [12]). At typical warm or hot extrusion temperatures, if remaining below 700 °C, the texture development during mechanical processing in the α phase of the current Zr–2.5 wt% Nb alloy is driven primarily by dislocation slip, with twinning making a minor contribution [8]. For processing at higher temperatures in the $\alpha+\beta$ phase field, other mechanisms may play a role, such as β phase deformation [13] and texture inheritance from the β phase through the Burgers relationship [14].

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The effect of interstitial species on the critical resolved shear stresses of the various slip and twinning modes active in zirconium is not well understood, and some disagreement exists in the literature. Studies on the effect of interstitial species on the active deformation mechanisms in zirconium alloys have focused on interstitial concentrations far below that of industrially applied alloys (*e.g.*, 390 wppm oxygen [15]). The role of interstitial impurities on twinning, in particular, is not established. Basic twinning models [16] suggest that increased interstitial content will discourage twinning in HCP by restricting the availability of sites for the matrix atoms to shuffle during the process, similar to the case of BCC alloys [17]. In titanium alloys, increasing oxygen content from 1000 wppm to 2000 wppm was observed to fully suppress twinning, and greatly reduce the activity of $\langle c+a\rangle$ slip [3]. The lower activity of $\langle c+a\rangle$ is reported to be due to the elimination of cross-slip. It is possible that the suppression of twinning is due in part to the suppression of $\langle c+a\rangle$ slip, as $\langle c+a\rangle$ dislocations have been reported to be necessary to accommodate the propagation of twins in Ti [18]. In HCP metals such as Ti and Zr, dislocations of $\langle c+a\rangle$−type are expected to be sensitive to oxygen due to the dislocations' extended structure and likelihood of dissociation [18]. The suppression of twinning by interstitial oxygen is similar to the suppression of the $\alpha\rightarrow\omega$ transformation under shock loading of titanium [19] and zirconium [20], as both are shearing processes.

We present herein a study on the deformation behaviour of hot-extruded Zr–2.5 wt% Nb. Three alloying chemistries have been used to allow for the study of the role of interstitial atoms during deformation. Samples were prepared with one of the three different final heat treatments. Compression tests were performed along the extrusion axis. *In situ* neutron diffraction was used to monitor the development of interphase and intergranular strains during deformation. The large data set collected can be used to further our understanding of the basic deformation mechanisms in this alloy, and allows for the investigation of the role of alloy chemistry in the activation of slip and twinning deformation modes. The data set will allow for the further development of modelling approaches free of the constraints of a limited texture and intergranular stress range. This paper presents experimental results at both the macroscopic and microscopic scales from which the effect of ingot chemistry, heat treatment and texture on the deformation of Zr–2.5 wt% Nb can be readily observed.

2. Materials and experimental method

Three Zr–2.5Nb ingots were prepared for this study, each with different alloying chemistry. The chemical composition of each of the ingots is given in Table 1. Ingots were prepared and machined into 2.0 in diameter rod. The rods were quenched from 1015 °C (1050 °C for the highest O content sample to allow for the increased transus temperature), with a 40 min hold prior to quenching. Billets were then machined to 16.6 in and encased in copper and iron for extrusion (as is the industry standard). Extrusion was performed at 815 °C with an extrusion ratio of 10:1, followed by two cold-drawing passes with 15% reduction to produce a final sample diameter of 0.447 in (~11.35 mm).

The rods were then subject to one of two heat treatments. The first heat treatment (heat treatment A, or HTA) was an annealing treatment below the $\alpha+\beta\longrightarrow\beta$ transus. Rod sections were heated in a furnace to 700 °C and held for 1 h, followed by a furnace cool. This temperature is high enough to suppress the formation of ω grains in the β phase [21], and will result in recovery of cold work and grain growth [22]. The second heat treatment (HTB) was performed above the $\alpha+\beta\longrightarrow\beta$ transus, and a phase transformation is expected [23]. Rod sections were heated in the furnace to 1100 °C, held for 30 min, cooled to 650 °C, held for 3 h, and furnace cooled to room temperature. The hold at 650 °C intended to relieve any stress resulting from the phase transformation and promote an $\alpha+\beta$ microstructure. During both heat treatments, the rod sections were contained in stainless steel bags that had been flushed with argon prior to sealing to prevent oxygen ingress. The rods were then machined to produce compression samples of 9 mm diameter and 18 mm length. Samples were also prepared in the as-received (*i.e.* hot extruded) state.

In situ compression tests were performed using the time-of-flight diffractometer at the ENGIN-X beamline at ISIS, Rutherford Appleton Laboratory. An in-depth description of the experimental setup for *in situ* loading has been published previously [24]. Samples are loaded at an angle of 45° to the incoming beam, allowing for measurement of diffraction peaks along two orthogonal directions. The measurement directions correspond to the loading axis and one direction perpendicular to the loading axis. Due to the orthonormal texture of the samples to be tested, the diffraction patterns produced along any direction normal to the loading axis are equivalent. Thus the lattice strains need to be only measured along two directions (in contrast to, *e.g.*, previous studies on rolled plate [25,26]). Samples were mounted in a computer-controlled hydraulic load frame, with a compressive holding stress of 5 MPa. This stress is necessary for the correct alignment of the samples, and has no practical influence on the lattice strain development during higher loads. An extensometer was attached to the sample across the midpoint to allow for the measurement of macroscopic strain during testing.

Tests were performed under stress control up to yield, and under position control during plastic deformation. This approach was taken to reduce the impact of creep during loading. After each incremental load was applied, the sample was allowed to relax from the peak load for 10 min, and then measurements were taken. The stresses and strains reported are the averages during the measurement period. Samples were tested to total macroscopic strains of 10%, and then unloaded to the holding stress of 5 MPa. Measurements were taken along the unload path to study the unloading behaviour and measure the final residual stresses.

Following compression testing, the crystallographic textures of the compression samples were measured using neutron diffraction at Chalk River Labs. Both pre- and post-deformation textures were measured. Pole figures were measured for the (0002), $(10\bar{1}0)$ and $(10\bar{1}1)$ lattice planes in the α-phase, with an angular resolution of 5°. Due to its low volume fraction, no pole figures were measured for the β-phase.

Table 1
Composition of the tested alloys. Values in wppm (except Nb, in wt%).

Alloy	O	Fe	H	Cl	C	Nb	Zr
Current	1176	547	< 3	< 1	71	2.66	Balance
High Fe	1320	1080	< 3	–	78	2.7	Balance
High O	3300	420	4	< 5	67	2.6	Balance

3. Data analysis

3.1. Lattice strain

In order to investigate the intergranular stresses developed during deformation, the results of the *in situ* neutron diffraction are displayed in terms of the strain on individual grain families. Each diffraction peak in the measured diffraction pattern