

Effect of hydrogenation conditions on the microstructure and mechanical properties of zirconium hydride



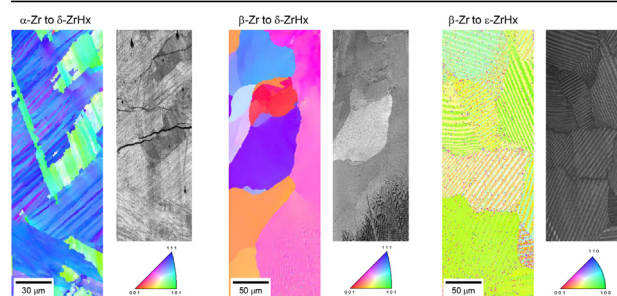
Hiroaki Muta ^{a,*}, Ryoji Nishikane ^a, Yusuke Ando ^a, Junji Matsunaga ^b, Kan Sakamoto ^b, Stefanus Harjo ^c, Takuro Kawasaki ^c, Yuji Ohishi ^a, Ken Kurosaki ^a, Shinsuke Yamanaka ^a

^a Graduate School of Engineering, Osaka University, Suita, Osaka 565-0871, Japan

^b Nippon Nuclear Fuel Development, Co. Ltd., Oarai-machi, Ibaraki 311-1313, Japan

^c Japan Atomic Energy Agency, Tokai-mura, Ibaraki 319-1195, Japan

GRAPHICAL ABSTRACT



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ABSTRACT

Precipitation of brittle zirconium hydrides deteriorate the fracture toughness of the fuel cladding tubes of light water reactor. Although the hydride embrittlement has been studied extensively, little is known about physical properties of the hydride due to the experimental difficulties. In the present study, to elucidate relationship between mechanical properties and microstructure, two δ -phase zirconium hydrides and one ϵ -phase zirconium hydride were carefully fabricated considering volume changes at the metal-to-hydride transformation. The δ -hydride that was fabricated from α -zirconium exhibits numerous inner cracks due to the large volume change. Analyses of the neutron diffraction pattern and electron backscatter diffraction (EBSD) data show that the sample displays significant stacking faults in the $\{111\}$ plane and in the pseudo-layered microstructure. On the other hand, the δ -hydride sample fabricated from β -zirconium at a higher temperature displays equiaxed grains and no cracks. The strong crystal orientation dependence of mechanical properties were confirmed by indentation test and EBSD observation. The δ -hydride hydrogenated from α -zirconium displays a lower Young's modulus than that prepared from β -zirconium. The difference is attributed to stacking faults within the $\{111\}$ plane, for which the Young's modulus exhibits the highest value in the perpendicular direction. The strong influence of the crystal orientation and dislocation density on the mechanical properties should be considered when evaluating hydride precipitates in nuclear fuel cladding.

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* Corresponding author.

E-mail address: muta@see.eng.osaka-u.ac.jp (H. Muta).

1. Introduction

Zirconium hydride is known to precipitate in the nuclear fuel cladding of light water reactors as a result of a reaction between the zirconium alloy cladding and the coolant water [1,2]. There are three well-known stable zirconium hydrides: γ -ZrH, δ -ZrH_x ($1.5 < x < 1.7$), and ϵ -ZrH_x ($1.8 < x < 2.0$). γ -ZrH displays a face-centered tetragonal (fct) structure with $a/c < 1$ and is regarded as a precursor of δ -ZrH_x. δ -ZrH_x has a face-centered cubic (fcc) structure and is the predominant hydride observed in the cladding material. ϵ -ZrH_x has an fct structure with $a/c > 1$ and is slightly distorted from the fcc structure of δ -ZrH_x. The formation of hydrides in α -zirconium displaying hexagonal-close packing (hcp) has been studied extensively over the last few decades [3–5] because these hydrides are brittle and significantly decrease the fracture toughness and elongation of the cladding. The hydride brittleness is considered one of the main factors of cladding deterioration. The δ -hydride has several crystal habit planes with the α -zirconium matrix and is observed to precipitate on the {10-10} prism plane, the {10-12} twinning plane, the {10-11} pyramidal plane, the {10-17} plane, and the {0001} basal plane. Among them, the {111} δ //{0001} α [6–9] is mostly observed in cladding. Thus, the cladding texture must be carefully controlled during the fabrication process. The cold working and heat treatment conditions are typically optimized for each cladding alloy, and the $\langle 0001 \rangle$ direction of the α -zirconium matrix grain is oriented in the radial direction of the cladding [2,10]. Consequently, the majority of the δ -hydride precipitates lie perpendicular to the radial direction of the cladding, which significantly reduces susceptibility of cladding to hydride embrittlement [11]. The formation, growth, and distribution of the hydride are paramount to the mechanical properties and robustness of light water reactor cladding.

Zirconium hydride has also attracted attention as a neutron moderator or fuel matrix in fast breeder nuclear reactors [12–14], a minor actinide transmutation target fuel in accelerator driven systems [15], and a neutron shielding material in fusion reactors [16,17]. The wide interest in zirconium hydride materials originates from the low thermal neutron absorption of zirconium and the high density of hydrogen when in the form of hydrides. The density is higher than those for pure liquid hydrogen and water. This high density allows the size of the neutron moderator or shielding material to be reduced while taking advantage of hydrogen's high neutron moderating and shielding ability due to its similar atomic mass to neutrons. Using the hydride components of these materials enables compact designs to be implemented in nuclear reactor and accelerator systems. Zirconium alloys and hydrides are thus indispensable materials in nuclear energy systems.

Although these hydrides' mechanical properties are primarily important to estimate the hydrogen embrittlement and their suitability for the various applications, only few attempts have so far been made to determine their properties. The reason is attributed to difficulty of the fabrication of bulk zirconium hydride samples. The fabrication of the samples without cracking is challenging due to the large volume change during hydrogenation and the brittleness of the hydride itself. The authors successfully fabricated samples by the hydrogenation of β -zirconium metal at high temperatures and investigated their mechanical properties [18,19]. However, the hydride precipitates in nuclear cladding are transformed from the α -zirconium metal and thus display an altered microstructure due to the large volume change. In addition, the reported mechanical properties of the δ -hydride vary significantly. For example, its Young's modulus is reported to be from 60 to 110 GPa ($\nu = 0.3$) [20] to 132 GPa [19]. The reason for this large variation in reported values has not yet been clarified. In the present study, to characterize the microstructure and its effect on the

mechanical properties of the bulk zirconium hydride, samples were fabricated using different hydrogenation conditions with consideration of the volume change at the metal-to-hydride transformation. The microstructure was investigated by neutron diffraction and electron backscatter diffraction (EBSD). The microstructure-dependent mechanical properties were measured by combination of indentation tests and EBSD observation.

2. Experimental

2.1. Sample preparation

Two δ -phase zirconium hydride samples, δ -ZrH1.66(α) and δ -ZrH1.66(β), and one ϵ -phase zirconium hydride sample, ϵ -ZrH1.90(β), were fabricated from zirconium metal (99.9% purity, <70 ppm hafnium, Rare Metallic Co., Japan) by hydrogen gas absorption in a modified Sieverts apparatus. (α) and (β) refer to the zirconium phase at the hydrogenation. δ -ZrH1.66(α) was hydrogenated from α -zirconium, δ -ZrH1.66(β) and ϵ -ZrH1.90(β) were hydrogenated from β -zirconium, respectively. The zirconium metal was arc melted, and the ingots were heated at 1073 K for 6 h at a vacuum of 10^{-5} Pa before hydrogenation. δ -ZrH1.66(α) was hydrogenated at 773 K and the others at 1073 K. The hydrogenation time was maintained at 12 h, and most of the hydrogen was absorbed using this procedure. The ingot was then cooled to 573 K at a rate of 0.25 K/min. Pure hydrogen gas (purity:7N) was used and H₂O impurity was removed at the gas introducing system cooled by liquid nitrogen. Fig. 1 shows a zirconium-hydrogen binary phase diagram with a normalized volume expressed along the z-axis. The volume of 1 M α -zirconium at 300 K was normalized to one. Literature data for the thermal expansion and hydrogen concentration dependence of the lattice constant [21–27] were used for the estimation. The transformation from α -zirconium to the δ -hydride at 773 K results in a volume change of approximately 20% [28]. Due to the large volume change, the formed δ -ZrH1.66(α) structure displayed many inner cracks. In contrast, the volume change arising during the transformation from the β -zirconium at 1073 K is moderate and is less than 10%. In addition, the zirconium matrix softened at high temperatures, easily deformed and accommodated the volume change caused by the hydride

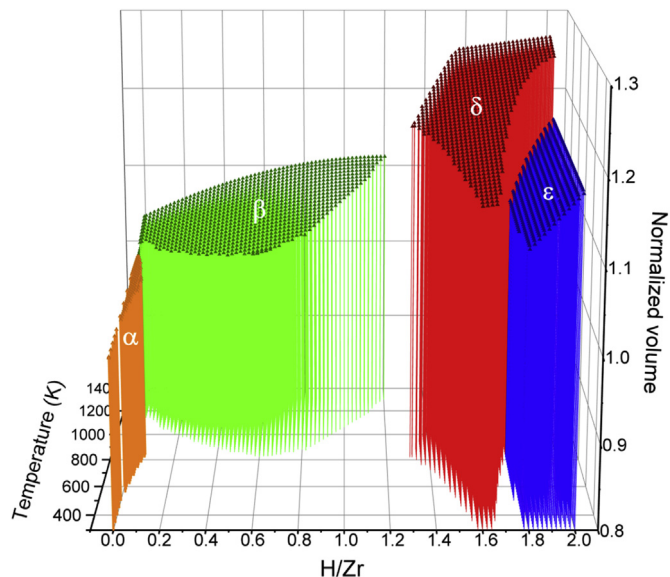


Fig. 1. Zr-H binary phase diagram. The z-axis represents the normalized volume of 1 mol of zirconium.

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