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Preventing cracking of nuclear fuel cladding

Question: *What mechanism controls the formation of zirconium hydride and how can it be restricted?*

Introduction

The ability of fuel cladding materials to maintain their mechanical properties over time is critical to the lifetime of nuclear reactors. Zirconium alloys are used for this application because of their compatibility with fuel materials, strength and ductility at reactor operating temperatures and corrosion resistance. However, while their corrosion resistance is good, it is not ideal.

Over time, the fuel cladding absorbs hydrogen through a corrosion reaction with the coolant water. Once the hydrogen level exceeds its solid solubility in the alloy, Zr hydride is precipitated, bringing about a marked reduction in the ductility of the alloy. Crack

growth typically occurs by fracture of hydrides and radially-oriented hydrides are much more detrimental than circumferentially-oriented ones. Avoiding premature cracking in fuel cladding therefore requires a sound understanding of the mechanisms that control the formation and fracture of the hydrides.

The aim of this work was to investigate the crystallographic relationships between different types of hydride and the zirconium alloy matrix and to draw conclusions about the mechanisms that control hydride formation.

EBSD Analysis

Orientation maps were collected using EBSD at three different positions on the radial-circumferential cross-section of a Zircaloy cladding tube.

Table 1:

Details of the experimental conditions.

Sample Preparation: Electropolished in 20% HClO_4 , 80% $\text{C}_2\text{H}_5\text{OH}$, followed by ion milling.

SEM Type: FEG-SEM

EBSD System: HKL CHANNEL5 with Nordlys II detector

Accelerating voltage: 20 kV

Probe Current: 1.5 nA

Results

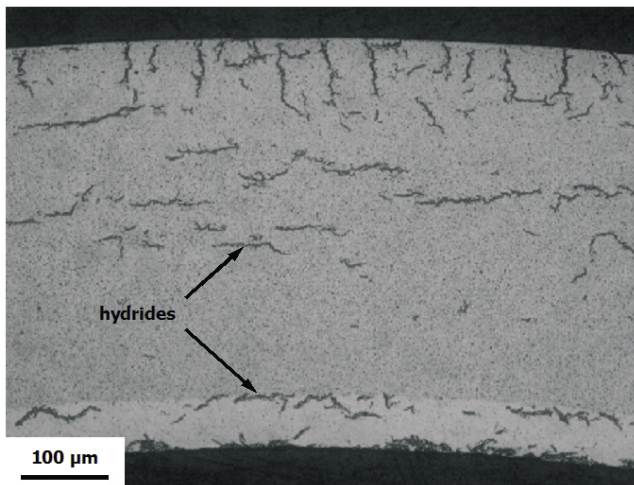


Figure 1:
 Optical micrograph of hydrogenated Zircaloy-2 cladding sample.

Figure 1 shows a cross-section of the zircaloy cladding tube. The hydrides are darkly etched. The tube can be thought of as consisting of three layers. The inner layer (the lighter grey layer at the bottom of the micrograph) is the zirconium liner. It contains mainly circumferentially-oriented hydrides. The middle layer also contains mainly circumferentially-oriented hydrides but most of the hydrides lie in the outer part of this layer (away from the Zr liner). The outer layer (at the top of the micrograph) contains mainly radially-oriented hydrides. There is no distinct boundary between the middle and outer layers.

EBSD maps were taken on each of the three layers to measure their textures and ascertain the orientation relationships between the hydrides and the zirconium or zircaloy matrix. The textures are shown using {0001} pole figures (Figure 2) and quantified using Kern's factors (Table 2). The middle and outer regions have virtually the same texture with 60% of basal poles oriented in the radial direction, 36% in the circumferential direction and only 4% in the axial direction (Table 2). The texture of the inner region (the Zr liner) is similar, except that there is less difference between the radial and circumferential basal pole fractions.

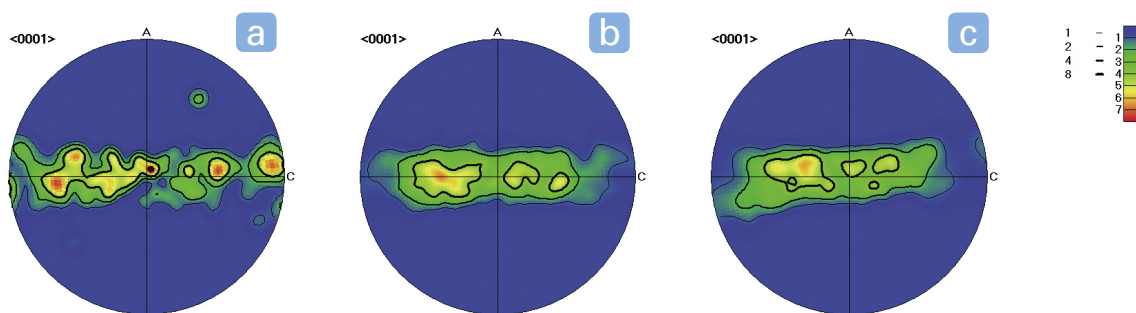


Figure 2:
 {0001} pole figures for (a) the inner layer of Zr, (b) the middle layer of zircaloy and (c) the outer layer of zircaloy. A marks the axial direction and C marks the circumferential direction.

¹ The Kern's factors describe the fractions of basal poles oriented in the radial (f_r), circumferential (f_c) and axial (f_a) directions, according to the formula $J_x = \sum V_i \cos^2 \alpha_i$, where i is the crystallite number, V_i is the volume fraction of crystallite i and α_i is the angle between the [0001] direction of the crystallite and direction x .

Table 2:

List of phases investigated in the DSS alloy using EBSD.

Region	f_r	f_c	f_a
Inner (Zr)	0.518	0.439	0.042
Middle (Zircaloy)	0.600	0.363	0.037
Outer (Zircaloy)	0.595	0.362	0.043

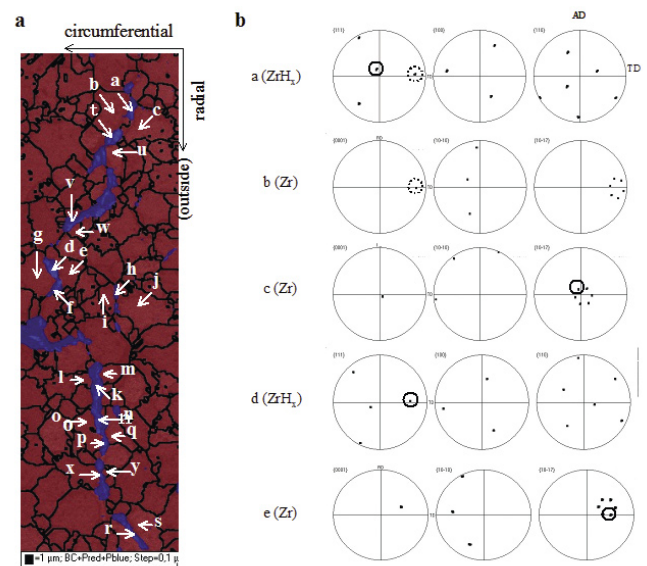


Figure 3:

shows a map taken on the outer layer where the hydrides are oriented radially. Pole figures for some points on the map are also shown. Two different orientation relationships were observed:

$$(0001)_{Zr} \parallel \{111\}_{ZrH_x}$$

$$\{10\bar{1}7\}_{Zr} \parallel \{111\}_{ZrH_x}$$

Figure 4 shows the dominant orientation relationship is $(0001)_{Zr} \parallel \{111\}_{ZrH_x}$ throughout the Zircaloy cladding tube. The $\{10\bar{1}7\}_{Zr} \parallel \{111\}_{ZrH_x}$ relationship with the matrix, observed for some hydrides in the outer layer, is not present in either the inner or middle layers. Further analysis of the data [1] showed that the $(0001)_{Zr} \parallel \{111\}_{ZrH_x}$ relationship is observed for both intra- and inter-granular hydrides, while the $\{10\bar{1}7\}_{Zr} \parallel \{111\}_{ZrH_x}$ relationship is observed only for inter-granular hydrides. It was also confirmed [1] that the outer region contains a much higher proportion of inter-granular radial hydride (82%) compared with the middle region (29%).

From these results, it was determined that the main change on application of a tensile stress is a strong tendency towards inter-granular precipitation on grain boundary faces perpendicular to the tensile stress axis. There is also intra-granular precipitation in grains with circumferential basal pole textures (where the habit plane is also perpendicular to the tensile stress axis). These two mechanisms give the high proportion of radially-oriented hydride observed in the outer region of the zircaloy. Without stress, the hydrides tend to precipitate on the basal planes and consequently most have a circumferential alignment due to the dominant radial basal pole texture.

The results support a mechanism of hydride formation that involves the effect of elastic strain energy on the terminal solid solubility of hydrogen [1]. They also indicate that strengthening the radial texture of the zircaloy will reduce the formation of radial hydrides, which are detrimental to ductility [1, 2].

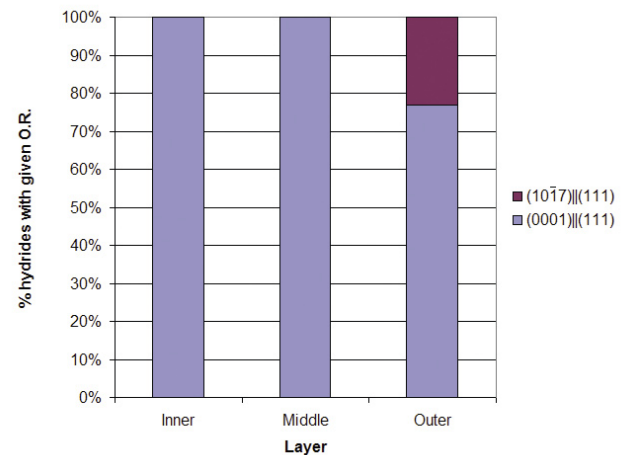


Figure 4:

Graph of percentage of hydrides in each layer of the tube having each crystallographic relationship.

Conclusion

The analysis established that the change in precipitation behaviour caused by application of a tensile stress is mainly a change in the location and orientation of the precipitates, not a major change in the crystallographic relationship between the matrix and the precipitates. The nature of the change in precipitation

provides support for a particular mechanism of hydride formation and suggests that susceptibility to cracking may be reduced by changing the processing so that the radial texture of the zircaloy is strengthened.

Answer: *The mechanism of hydride formation involves the effect of elastic strain energy on the terminal solid solubility of hydrogen. Strengthening the radial texture of the zircaloy should restrict the formation of detrimental radial hydrides.*

References

1. Une, K., Nogita, K., Ishimoto, S. and Ogata, K., J. Nuc. Sci. Tech., 2004, 41, 731.
2. Kearns, J. J. and Woods, C. R., J. Nucl. Mater., 1966, 20, 241.

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