Microstructural and Microtextural Studies in Zr Based Alloys

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Abstract

An overview of microstructural and textural changes taking place during the processing of the Zirconium alloys and their role in the hydriding behavior is presented. The linkage between the microstructural evolution and textural development is demonstrated in case of Zircaloy-4 clad tube fabrication. An algorithm is presented for the reconstruction of the high temperature phase microstructure, using which the significant variant selection taking place during the transformation of β-Zr into α-Zr is demonstrated. The role of microstructure on the hydride formation is discussed in terms of effect of grain/phase boundaries.

Introduction

Some of the most functionally critical in-reactor components like pressure tube and clad tubes are made up of Zr based alloys. The service performance of the components made by these alloys largely depends on their as-fabricated microstructure and texture. Hydride formation, irradiation creep and growth are some of the important life limiting phenomena which are directly influenced by the component’s microstructural and textural conditions. Hence it is important to understand (a) the evolution of the microstructure and texture during the fabrication (b) the role of the microstructure on the phenomena like hydride formation etc [1]. Such understanding helps in tailoring the microstructures for mitigating the ill effects of the hydride formation. Understanding of the microstructural evolution as a function of process parameters also helps in optimizing the fabrication flow sheet to obtain the desired microstructures that enhance component performance.

The present paper presents an overview of microstructural and textural evolution studies in various Zr based alloys and the microtextural aspects of hydride formation in them. Along with the conventional characterization tools of transmission electron microscopy (TEM), and X-Ray Diffractometry (XRD), scanning electron microscopy (SEM) based electron back scatter diffraction (EBSD) has also been extensively used. EBSD has the advantage of offering images with reasonably higher spatial resolution combined with the local orientation information. Thus, it combines the imaging capabilities of SEM with the textural capabilities of XRD [2] and opens up some interesting possibilities which are demonstrated in this article.

Evolution of Texture and Microtexture during Zircaloy-4 clad fabrication:

Thermo-mechanical processing (TMP) steps used for the fabrication of Zr components are aimed at obtaining required dimensional tolerances and optimized microstructure. The final microstructure, and associated properties, is often a cumulative outcome of series of changes occurring at each stage of processing. Hence, a systematic characterization of the microstructural and textural evolution in each of the stages of the PHWR clad fabrication was carried out and presented in Fig.1. The fundamental mechanisms responsible for observed changes were investigated and the important observations emerging out of this study could be summarized as below.
• β quenching produced a nearly randomized texture and relatively large Widmanstatten grains of a phase (Fig 1b).

• Hot extrusion (HE) had resulted in the formation of the bi-modal grain size distribution and the development of a strong crystallographic texture (see Fig 1b). The HE texture was essentially made up of two fibers viz, ND || (0001) and ND || (1010) which are characteristic fibers expected of recrystallization and deformation respectively.

• Subsequent cold deformation steps have resulted in strengthening of the (1010) fiber while the annealing steps made the (0001) fiber stronger (see the orientation distribution function (ODF) maps in the Fig 1c). Microtexture analysis had revealed that the observed textural changes were essentially from grains of larger size.

• TEM investigations have revealed the signatures of the dynamic recrystallization (formation of high angle grain boundaries, and equi-axed grains with relatively low dislocation density in conjunction with elongated grains) during the HE. Pilgered structures on the other hand exhibited heterogeneous microstructures with heavy dislocation networks (Fig.1d). TEM of “Final annealed” sample indicated that the sample had undergone partial recovery.

Reconstruction of high temperature β phase:

As shown in the previous section one of the important steps of the component fabrication is the β-quenching step which results in texture randomization. However, the degree of texture...
randomization and the resulting microstructure subsequent to quenching are functions of the nature of the phase transformation that occur during quenching and the prior high temperature microstructure. Hence, knowledge of microstructure in the high temperature phase regime and nature of transformation in terms of variant selection can be helpful in optimizing the quenching parameters. In this context, we had developed an algorithm to reconstruct the microstructure of the high temperature phase using the micro-texture data (EBSD data) of the room temperature phase [3].

The principle of reconstruction is based on the fact that in case of Burger’s orientation relationship (OR), each a grain (product) can have 6 possible β (parent) variants. Hence by considering at least three a grains which have formed form same parent β grain it is possible to deduce the orientation of the parent grain and thus reconstruct the parent microstructure. The actual algorithm is explained below.

1. Consider a product grain Gi (where i = 1, 2, 3...) has G1, G2, ..., Gn as its neighbors – see Fig. 2a. Fig. 2b shows possible triplets containing Gi and its neighbors with the condition that each grain of the triplet is neighbor to other two.

2. For grains in a given triplet find all possible parent orientations

\[ B^β_i = T^α_i \rightarrow β G^α_i \quad B^β_i = T^α_i \rightarrow β G^α_i \]  \hspace{1cm} (1)

Based on the above algorithm, we reconstructed various transformed microstructures in case of Zircaloy-4 samples which have undergone a wide range of transformations ranging from diffusional to martensitic. An example of such reconstructed microstructure is presented in the Fig 3, which also shows the identification of the product variants based on the just mentioned algorithm. It is clear that there is considerable degree of variant selection (see Fig. 3d).

Where, \( T_1^{α_i \rightarrow β} \) and \( S_{m,k,l}^{α_i,β} \) are the hexagonal symmetry operators D is the matrix representing Burgers OR for \( β \rightarrow α \) and \( i = 1..3 \) are the grain IDs in the selected triplet; \( B^β \) is the parent orientation for the product orientation of \( G^α \). Of these parent solutions, common solution to all the three grains of the triplet is selected as a ‘potential solution’.

3. These steps are repeated for all triplets of Gi yielding, say, \( n \) potential solutions for Gi. In order to assign an ‘optimum’ solution to Gi, we find the mean solution \( (S_{m,k,l}) \) and mis-orientation \( (ΔS_{m,k,l}) \) between each pair of the potential solutions: \( S_k \) and \( S_l \) where \( k \) and \( l \) run from 1 to \( n \).

\[ ΔS_{m,k,l} = \cos \left( \frac{\text{trace}(S_k S_l)}{2} \right) \]  \hspace{1cm} (2)

Reject any \( S_{m,k,l} \) for which \( ΔS_{m,k,l} \) is more than user specified maximum misorientation tolerance \( (δ_{max}) \). Final solution \( S_{final} \) for Gi is the mean of all \( S_{m,k,l} \) weighted by \( W_{k,l} \) (weightage factor for \( S_{m,k,l} \)).

\[ S_{final} = \frac{\sum S_{m,k,l} W_{k,l}}{\sum W_{k,l}} \]  \hspace{1cm} (3)

Fig. 2: Schematic showing the product a grains. Grains belonging to common parent b are marked with the same color/shade. Subscripts (G) indicate the product grain IDs. (a) Adopted algorithm. Let \( G_i = 5 \). Neighbors of \( G_5 \) are \( G_1, G_2, G_4, G_7 \) and \( G_8 \). The triplets formed by \( G_5 \) are \[ \{5,1,2\}, \{5,2,8\}, \{5,8,7\}, \{5,7,4\}, \{5,4,1\} \]. Note that all the three grains of these triplets are neighbors to each other. Triplets either have a common parent variant, i.e. a “potential” solution for the parent b, or “no solution”. The next step is to link the “potential” solutions through a generalized misorientation criterion.
Based on the above algorithm, we reconstructed various transformed microstructures in case of Zircaloy-4 samples which have undergone a wide range of transformations ranging from diffusional to martensitic. An example of such reconstructed microstructure is presented in the Fig 3, which also shows the identification of the product variants based on the just mentioned algorithm. It is clear that there is considerable degree of variant selection (see Fig. 3d).

Some of the important advantages of this algorithm in comparison to other existing ones are its (a) less sensitivity to measurement errors in orientation measurement (b) practical elimination of spurious unification of the grains belonging to different parent grains (c) independence of calculated solution from user defined angular tolerance ($\delta_{\text{max}}$).

Thus, we are able to reconstruct the high temperature microstructure, and correlate the observed textural developments by identifying the variant selection taking place during the phase transformation using the just mentioned algorithm. This capability is expected to be of immense help in optimizing the $\beta$ quenching process and gaining further insight into the mechanism of $\beta$ to $\alpha$ phase transformation.

**Role of grain/phase boundaries in hydride formation:**

Hydride formation is known to be one of the life limiting factors in case of Zr based structural components used in thermal nuclear reactors. Understanding the mechanism of hydride formation can help tailor suitable microstructures for better hydride mitigation in these alloy components. Previous works, which are largely based on TEM and optical investigations, have indicated that the hydrides preferentially form along the grain boundaries of the matrix phase. However, very little information was available on the role of nature of the grain boundaries and interfaces in controlling the hydride formation, as these techniques inherently lack local orientation information and suffer from poor statistics [4,5].

In the present article we report, substantial improvement in understanding of the role of grain/phase boundaries with explicit use of local orientation measurements through EBSD technique.

Fig. 4: Hydride distribution in Zirconium based alloys (a) Zircaloy-2 (b) Zr-2.5%Nb alloy. It is clear that in both the cases the hydride phase is along the interfaces of the grains only.
• In case of single phase alloy, (Zircaloy-2), formation of hydrides was preferred on certain grain boundaries. Those boundaries which are characterized by low coincident site lattice (CSL) values, in general, were resistant to hydride formation. This could be attributed to relatively higher atomic order among such boundaries which lowers their energy and thus decreases their potency for heterogeneous nucleation of secondary phases.

• Higher resistance to hydride formation was also observed in boundaries surrounded by elastically harder grains. This observation was rationalized by the expected higher stresses generated during the hydride formation (due to inherent volumetric expansion involved in hydride formation) along boundaries formed elastically hard grains.

• Hydrides in the two phase Zr-2.5%Nb alloy in a completely recrystallized condition have formed primarily along α/β interfaces, with only a minor fraction of hydrides being along a/a grain boundaries.

• Majority of the hydrides were along those α/β interfaces which had a misorientation corresponding to burger’s OR. However, not all the α/β interface which are related by the burger’s OR are having the hydrides along them. Selection α/β interfaces as the favorable nucleation sites by the hydrides was attributed to high hydrogen partitioning between the α and β phases on account of the large differences in solid solubilities of hydrogen in α and β phases. This makes α/β interface the nearest available heterogeneous nucleation site for most the precipitation of hydride.

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References