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Formation of very large 'blocky alpha' grains in Zircaloy-4

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ABSTRACT

Understanding microstructure and its evolution is very important in safety critical components such as cladding in nuclear reactors. Zirconium alloys are used as cladding materials due to their low neutron capture cross section, good mechanical properties and reasonable corrosion resistance. These properties are optimised, including grain size and texture control, to maximise performance in thin (<1 mm wall thickness) tubes in water reactors. Here we show that very large grains (>0.5 mm) can be generated systematically during controlled deformation and subsequent heat treatments. We observe that the texture of these grains is controlled either by twinning or prior texture, depending on the strain path. Their nucleation, growth and texture can be controlled through strain path and deformation level. This work provides detailed understanding of the formation of these very large grains in Zircaloy-4, and also opens up opportunities for large single crystal fabrication for mm scale mechanical testing.

1. Introduction

Zircaloy-4 is dilute zirconium alloy used in nuclear power applications as fuel rod cladding due to its low neutron capture cross-section and good mechanical strength. At room temperature it is hexagonal close-packed (HCP) α phase, with a 0.5 % volume fraction of second phase particles around 100 nm in diameter [1]. The nominal chemical composition of Zircaloy-4 is Zr − 1.5 wt%Sn − 0.2 wt%Fe − 0.1 wt%Cr [2].

A typical Zircaloy-4 fuel tube in a pressurised water reactor has a wall thickness of 0.57 mm [3]. Tube walls are thin to minimise neutron absorption and maximise fuel efficiency, but need to withstand high stresses during operation. A fine, uniform grain size is desirable in order to optimise strength, minimise stresses from thermal expansion and irradiation, and ensure a relatively homogeneous strain state along the entire fuel tube. Understanding the evolution of grain size during service is important for component life estimations, and excursions of grain size towards very large grains, the so called 'blocky alpha' structure (grains >300 μm with irregular and wavy grain boundaries) [4], need to be understood.

Since the tube walls are thin, blocky alpha grains could span the entire width of a fuel tube wall. These very large grains can cause issues, since zirconium is anisotropic due to its HCP crystal structure [5]. The texture spike from a single large grain can affect anisotropic material properties such as yield [6] and thermal expansion [7]. An absence of grain boundaries can impact properties such as strength (as small grains result in a stronger product [6]) and change ageing regimes such as irradiation growth and creep [8]. Furthermore, the orientation of the blocky grains can affect degradation mechanisms such as hydride embrittlement, if the grain is poorly oriented for brittle hydride plates to form on near basal planes [9] or reorient along the principal stress direction [10], which is often a compressive radial stress for fuel cladding tubes [11].

This paper explores the formation of blocky alpha grains in Zircaloy-4. First, some terms relating to recrystallisation and grain growth processes will be defined. In the results section, observations of blocky alpha formation via strain-anneal processing using both uniaxial compression and three point bending geometries are reported. In the discussion section, a mechanism for blocky alpha growth and orientation selection during nucleation is proposed.

2. Grain growth and recrystallisation processes

Recrystallisation is the formation of a new grain structure in a deformed material by migration of high angle grain boundaries (>10 − 15°) to reduce stored strain energy. In plastically deformed materials the energy from plastic work is eliminated by nucleation and growth of new grains via **primary recrystallisation** [12].

Grain growth can occur on further annealing after recrystallisation. It is the migration of high angle grain boundaries where the

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driving force for grain boundary migration is the reduction of grain boundary interfacial energy. In **normal grain growth** the smallest grains shrink and are consumed by neighbours, so that the average grain size increases. Normal grain growth is a continuous transformation, which means that it occurs homogeneously and simultaneously throughout the parent structure [12]. **Abnormal grain growth** occurs if normal grain growth is supressed, e.g. by pinning from second phase particles. A minority of grains grow rapidly and consume neighbouring grains. This leads to a bimodal grain size distribution until all the initial grains are consumed, then the grain size distribution is once again unimodal, with a much larger average grain size than the starting material. Abnormal grain growth is also known as secondary recrystallisation [13]. The driving force for both normal and abnormal grain growth is the reduction of grain boundary area [12].

Abnormal grain growth and primary recrystallisation are discontinuous transformations. In these transformations, there is a sharp interface between transformed and untransformed material which sweeps through the material as the transformation proceeds [12]. Discontinuous transformations generally are also termed 'nucleation and growth' transformations as these processes can be divided into two steps: the formation of a stable nucleus which is energetically favourable to grow, and then growth of that nucleus. For example, in primary recrystallisation, the nuclei are usually recovered subgrains; in abnormal grain growth, the nuclei are the pre-existing recrystallised grains [12].

Nucleation site limited primary recrystallisation [14], also known as 'abnormal' recrystallisation [15], is a recrystallisation phenomenon which can produce very large grains. Similarly to abnormal grain growth, a minority of grains rapidly consume neighbouring grains to form a very large final grain structure. 'Abnormal' and primary recrystallisation are mechanistically indistinct, and the difference in transformed grain size is due to the extreme sparseness of nuclei during recrystallisation.

Although abnormal grain growth and (nucleation site limited) primary recrystallisation can produce similar microstructures, the transformation driving force is different between them: the driving force for nucleation site limited primary recrystallisation is the lowering of stored strain energy in the material, whereas the driving force for abnormal grain growth is the reduction of grain boundary area.

Abnormal grain growth and nucleation site limited primary recrystallisation can be distinguished if the transformation driving force can be isolated, as has been studied by Chen et al. in a friction stir welded aluminium alloy [16], where pre-annealing was used to recover the deformed structure before further heat treatment to produce large grains. In addition, often the speed of the transformation growth front in metals is one order of magnitude faster for recrystallisation (~10 μm/s) than for abnormal grain growth (<1 μm/s), so the mechanisms can be distinguished if typical transformation kinetics are known [15].

Strain induced grain boundary migration (SIBM) is a nucleation mechanism for recrystallisation. A pre-existing grain boundary segment bulges into an adjacent grain, the driving force for which is to reduce local stored energy ahead of the bulging grain boundary segment. The stored energy can take the form of either residual elastic energy or a higher dislocation density [12,15,17]. SIBM provides a mechanism for a recrystallised grain to nucleate without the formation of new recrystallisation nuclei from e.g. recovered subgrains; hence, it is regarded as a nucleation process [15,18].

In SIBM, the bulging grain boundary migrates away from its centre of curvature, and increases the surface energy and line tension of the grain boundary. For SIBM to be energetically favourable, the volume ahead of the migrating grain boundary must have higher stored energy which can be swept away by the migrating grain boundary [18]. The stabilisation of the bulging grain boundary segment is the limiting step in SIBM, leading to an incubation period in transformations which nucleate via SIBM [15].

The strain-free material created from the sweeping of the grain boundary takes on the orientation of the pre-existing grain behind the sweeping boundary segment. As a result, the recrystallised textures are generally similar to, or a subset of, the deformed textures [18]. This is not always true for primary recrystallisation via subgrain growth, where entirely different textures can form, such as in heavily deformed copper [12].

2.1. Prior observations of recrystallisation and grain growth in Zr

2.1.1. Primary recrystallisation and normal grain growth

Primary recrystallisation temperatures in reactor grade pure Zr range between 550 and 700°C after 10 − 50 % strain, and the recrystallisation temperature decreases with increasing cold work [19]. A finer grain size can be achieved by more cold work. Normal grain growth after primary recrystallisation is extremely limited.

Jedrychowski et al. [20] showed that recrystallisation in moderately (17 %) strained, textured commercially pure Zr (α-Zr) is likely to be via SIBM. SIBM is the bulging of a boundary segment ahead of a nearly strain-free subgrain into highly deformed grains in the heterogeneously deformed structure. It allows recrystallisation to nucleate without the formation of a recrystallisation nucleus, which has high formation energy. By comparing experimental textures with Monte Carlo Potts models, they showed that the SIBM model describes the texture evolution more closely than classical nucleation and growth models. This result is independent of texture component strained, which is important as texture has a strong influence on deformation systems activated in Zr.

2.1.2. Abnormal grain growth

Literature reports of abnormal grain growth in Zr are generally based on observations of very large grains in the final microstructure. This section is reported according to the conclusions of the cited literature, but it should be noted that similar grain structures can arise from either abnormal grain growth or nucleation site limited primary recrystallisation, even though the driving forces for these mechanisms are different.

2.1.2.1. Effect of prior strain. Washburn [21] observed significant grain growth in Zircaloy-4 plate after deforming in tension to strains between 2 % and 12 %, then annealing for different times at 800°C (Fig. 1(a)). Reducing the temperature to 700°C resulted in a suppression of grain growth, and after 167 h (10^4 min) some samples had grown slightly to an average grain size of around 50 μm (0.05 mm) (Fig. 1(b)). Annealing at 500 and 600°C produced no grain growth for up to 167 h (10^4 min) at strains between 2 % and 12 %.

Schemel [22] observed abnormal grain growth during annealing after 5–8 % cold reduction of zirconium alloys. This was reported to be common after straightening or forming operations.

Gray [19] observed abnormal grain growth (also called 'secondary recrystallisation') in reactor grade pure Zr. This was achieved by annealing 10 % and 50 % cold rolled Zr in 10^{-5}Pa vacuum at 800°C for 1000 min or longer. The largest grains were up to 2 mm in diameter, with wavy and irregular boundary traces between large grains. Small island grains, not consumed by the grain growth process, are also observed. The grain size increases with annealing time, with larger grains observed after four days than after one day. The 10 % cold rolled sample produced smaller grains than the 50 % cold rolled sample for the same annealing conditions.