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Investigating the thermal stability of irradiation-induced damage in a zirconium alloy with novel in situ techniques

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ABSTRACT

Zr alloys exhibit irradiation-induced growth and hardening which is associated with the defects and dislocation loops that form during irradiation. In this study, state-of-the-art in-situ synchrotron X-ray diffraction (SXRD) and transmission electron microscopy (TEM) techniques were used to investigate the stability of dislocation loops in two proton-irradiated Zr-Fe binary alloys in real time. Complementary data from both techniques show rapid annealing of a-loops occurs between 300 °C and 450 °C. Line profile analysis was performed on the SXRD patterns using the convoluted multiple whole profile analysis tool, to calculate the change in a-loop line density as a function of post-irradiation heat treatment temperature and time. At temperatures below 300 °C, no significant decrease in a-loop density was detected when held for 1 h at temperature. From this SXRD experiment, we calculate the effective activation energy for the annealing process as 0.46 eV. On-axis in-situ STEM imaging was used to directly observe a-loop mobility during heating cycles and confirm that a-loops begin to glide in the trace of the basal plane at ~200 °C in a thin foil specimen. Such a-loop gliding events, leading to annihilation at the foil's surfaces, became more frequent between 300 and 450 °C.

1. Introduction

The use of zirconium (Zr) alloys in nuclear reactor fuel assemblies is prevalent due to their low thermal neutron absorption cross-section and acceptable structural properties at reactor operating temperatures [1]. Zr alloys undergo irradiation-induced growth (IIG), one of the major life-limiting factors of the fuel assemblies, which is suggested to be due to the anisotropic diffusion of point defects generated by irradiation damage [2], and is correlated with the formation of dislocation loops [3–6]. There are two dominant types of loops that form during irradiation of Zr alloys, commonly referred to as a-loops and c-loops. The a-loops appear first at low dose and have a Burgers vector of $\frac{1}{3}\left\langle 11\bar{2}0\right\rangle$ and a habit plane of either the first or second order prismatic planes [7]. The appearance of c-loops is correlated to the onset of the accelerated growth regime, known as breakaway growth [8]. c-loops reside on the basal plane and have a Burgers vector of $\frac{1}{6}\left\langle 20\bar{2}3\right\rangle$ [9].

Irradiation results in a significant change to the mechanical properties of Zr alloys, primarily an increase in hardness [10–16]. It has been proposed that this change is due to the formation of dislocation loops and irradiation-induced precipitates, which act as barriers to dislocation movement during mechanical deformation [11,12,17,18]. The recovery of mechanical properties via heat treatments or temperature variations along a fuel assembly is therefore a key avenue of study for Zr alloys. Data generated from annealing experiments provide vital information regarding the stability of defects and may also provide insight into the mechanisms of irradiation damage. Further, it is important to predict the annealing behaviour of irradiated Zr alloys as residual heat during fuel waste storage could impact the structure of the irradiation-induced defects. For instance, it has been observed that the growth strain can be fully recovered in pre-breakaway neutron-irradiated Zircaloy-2 specimens through annealing [3]. It is also important to understand the effect temperature has on irradiated Zr alloys when considering ex-situ corrosion testing in a high temperature autoclave environment.

Previous annealing studies have been performed on neutron-

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irradiated Zr alloys that contained a high density of a-loops and exhibit irradiation-induced hardening [12,14,15,17]. These studies have shown that post-irradiation annealing leads to recovery of the base mechanical properties for alloys containing only a-loops [12,15,17]. This recovery has been correlated with an overall reduction in dislocation loop density and an increase in the average loop diameter [12,17,19]. Ribis et al. used microhardness values, transmission electron microscopy (TEM) observations and a cluster dynamics model to show that the recovery of the hardness to non-irradiated levels is correlated with a reduction in dislocation density [12].

To date, the majority of experimental work has been performed ex-situ providing only snap-shots of the microstructural and mechanical property evolution during the annealing process, using transmission electron microscopy (TEM) and mechanical testing techniques. The present work aims to demonstrate the annealing of proton-irradiated binary Zr-Fe alloys in real time using novel in-situ techniques. Specially fabricated Zr-Fe binary alloys have been used in this study to eliminate some of the complexity inherent in commercial alloy systems, whilst retaining key aspects of material evolution under irradiation, allowing the effect of specific microstructural features to be evaluated. Proton irradiation has previously been shown to be a good surrogate for neutron-irradiation in Zr alloys, providing similar damage structures and chemical evolution [10,20–23]. Proton irradiation allows for the design of systematic studies, achieving comparable damage levels in short timescales with low residual activity compared to neutron-irradiated samples.

As well as TEM, line profile analysis has been used to investigate the dislocations formed during irradiation in Zr alloys. The software package, extended Convoluted Multiple Whole Profile (CMWP) was originally developed to calculate the dislocation densities in cold-worked material but has also been used previously to calculate the dislocation densities in neutron-irradiated Zr-2.5Nb samples [24]. Seymour et al. used CMWP to calculate dislocation densities in neutron-irradiated Zircaloy-2 and found that despite some differences in absolute dislocation densities, the trends observed with CMWP, with respect to dislocation density changes with increasing damage level, were similar to those observed in TEM investigations [25]. In this work CMWP will be used to track the change in dislocation density during the in-situ SXRD annealing experiment.

The present study aims to investigate the stability of a-loops during heating using two complementary in situ techniques. In-situ synchrotron X-ray diffraction (SXRD) was used to obtain bulk information and in situ heating in a TEM was used in a complementary investigation into the mechanisms of recovery with superior spatial and temporal resolution. These novel techniques allow for new insight into the mechanisms of defect annealing in Zr alloys, allowing for determination of an accurate temperature for the onset of annealing and for an activation energy to be calculated, which will enable better informed selection of parameters for future experiments.

2. Experimental methods

2.1. Material

Fully recrystallised Zr-0.6Fe and Zr-0.1Fe (wt%) alloys, with the characteristic split basal texture [26], were proton-irradiated at the University of Michigan Ion Beam Laboratory's 1.7 MeV Tandetron acceleration facility, operated at 2 MeV. The samples were irradiated to a damage level of 3 dpa (3.415×10^{19} protons cm^{-2}) for the Zr-0.6Fe alloy and 1.5 dpa (1.708×10^{19} protons cm^{-2}) for the Zr-0.1Fe alloy, with an irradiation temperature of 350 ± 9 °C. The nominal dpa levels are extracted from the damage profile at a depth of 12 μm from the sample surface, as shown in Fig. 1. A current density of 0.2 μA mm^{-2} was maintained throughout the irradiation resulting in a damage rate of ~1.096×10^{-5} dpa s^{-1}. Therefore, the 3 dpa sample was irradiated for 76 h and the 1.5 dpa sample was irradiated for 38 h. The dose calculations were performed using the SRIM software package with the "Quick" Kinchin and Pease damage calculation [27] in pure Zr with a displacement threshold energy of 40eV. The predicted damage profile for the 1.5 dpa sample can be seen in Fig. 1, with the black vertical line indicating the region studied in the TEM, at a depth of ~12 μm. The dose level stated for the binary Zr-Fe alloys is based on the damage level at 40% of the Bragg peak from the sample surface in order to indicate the dose in the studied plateau region of the damage profile. The damage profile shows a large dependence on depth, and therefore it is important that the flatter region of the profile is studied in order to minimise variation in damage levels within a sample.

The binary 1.5 dpa Zr-0.1Fe alloy used in this work has been investigated in detail as part of another study, looking into the effect of redistributed Fe from SPPs on dislocation densities [20]. It was found that this alloy contains ~1–2 μm coarse Zr_3Fe secondary phase particles (SPPs) unevenly distributed throughout the sample. Close to these SPPs, which have undergone some irradiation-induced dissolution, Fe has been observed to segregate to pyramidal loops with a likely Burgers vector of $\frac{1}{3}\langle 11\bar{2}3 \rangle$ [20]. Away from the SPPs, the matrix and the dislocation structure is akin to that in pure Zr, with no Fe clustering observed [20]. The region investigated by TEM in this study was <1 μm away from an SPP and so contains very low levels of Fe in the matrix. Increasing the Fe wt% in the alloy results in an increased SPP volume fraction from <1% to ~5% between the Zr-0.1Fe and Zr-0.6Fe alloys. However, the SPPs in Zr-0.6Fe are significantly coarser than in Zr-0.1Fe and hence the inter-particle spacing remains similar. Therefore, the majority of the matrix in the sample studied using SXRD will not be affected by Fe redistribution from the Zr_3Fe SPPs and consequently, even though two variants of Zr-Fe binary alloys have been studied, the slight compositional differences should not affect the validity of comparisons between samples.

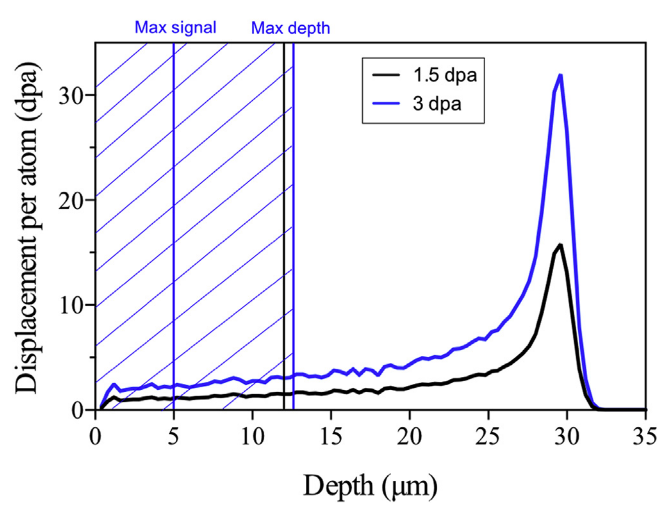

Fig. 1. Damage profiles generated using SRIM for pure Zr calculated using the "Quick" method. The simulation was carried out using 2 MeV protons and a displacement threshold energy of 40 eV for Zr. The calculation of dpa was carried out assuming a constant current density of 0.2 μA mm^{-2} with a damage rate of 1.3×10^{-5} dpa s^{-1}. The black vertical line represents the region studied using TEM (~12 μm from the sample surface), and the blue hatched area represents the region studied via SXRD. The maximum penetration of the X-rays and the depth from which the maximum intensities of diffracted X-rays originate are indicated. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)