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Evolution of the radiation-induced defect structure in 316 type stainless steel after post-irradiation annealing

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HIGHLIGHTS

- TEM study of irradiated CW316 steel after post-irradiation annealing.
- Frank loops were removed after annealing at 550 °C, by unfaulting and growing.
- The cavity density decreases after annealing at 550 °C, but not completely removed.
- Frank loop and cavity removal is controlled by the annealing temperature.
- The dissolution of γ' precipitates is controlled by the iron diffusion length.

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ABSTRACT

The thermal stability of Frank loops, black dots, cavities and γ' precipitates in an irradiated 316 stainless steel was studied by transmission electron microscopy. The samples were retrieved from a thimble tube irradiated at around 320 °C up to 80 dpa in a commercial nuclear power reactor, and thermally annealed, varying both annealing temperature and time. With increasing annealing temperature the density of all defects gradually decreased, resulting in the complete removal of Frank loops at 550 °C. In contrast to other defects, the density of the γ' precipitates sharply decreased with increasing annealing time, which indicates that the dissolution of the γ' precipitates is governed by the iron diffusion length.

1. Introduction

Irradiation assisted stress corrosion cracking (IASCC) is an important effect limiting the integrity of light water reactor (LWR) core components, especially with respect to long-term operations [1]. In stainless steels, IASCC occurs as an interplay of various degradation processes due to the material being exposed to elevated temperature, stress, intense neutron bombardment and corrosion [2–9]. The complexity of IASCC is illustrated by the fact that in boiling water reactors (BWR) the IASCC occurs after about 1 dpa of accumulated damage, while in pressurized water reactors (PWR) it occurs after 4 dpa. Evidently, both changes of the water environment and the microstructure of the irradiated alloy [3] are influencing IASCC, but their individual contributions and the importance of their mutual interplay are not yet fully understood.

In the framework of the life extension of nuclear reactors to 60 years and beyond, mitigation strategies against IASCC are considered. They are important for the safety as well as for maintenance cost reductions, even though IASCC may not be a lifetime limiting factor because most stainless steel components are replaceable. Hereto, the water chemistry could be optimized by adding Zn to reduce IASCC and Li to increase pH [10]. Alloy compositions could be adjusted to reduce susceptibility to IASCC. It is considered beneficial [1] to have high Ni and Cr contents, low Si content, absence of brittle oxide and nitride inclusions, high coincident site lattice fraction of grain boundaries, low connectivity of high-angle grain boundaries and grain boundary coverage by chromium.

As an alternative, one could consider to recover the initial mechanical properties by thermal annealing. Post-irradiation annealing (PIA) experiments have shown that both hardening and radiation-induced segregation (RIS) are reduced, where the radiation-induced hardening appears to recover faster than RIS [11]. On the other hand, annealing at high temperatures will promote helium bubble growth, specifically at grain boundaries and line dislocations [12,13]. Helium is produced as a result of (n,α) interactions. It is practically insoluble in metals and it will cluster and form bubbles at the radiation induced defects. Upon annealing at high temperature, the radiation induced defects are removed

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and He will form bubbles at locations of low free energy, like line dislocations or grain boundaries. Bubbles grow by migration and coalescence [14]. In 304-type stainless steel, bubbles become visible after annealing at 650 °C and reach a size of 1–4 nm [12]. PIA does not have a general healing effect for BWR conditions as the material is thermally sensitized during annealing, leading to cracking in high potential water. On the contrary, for PWR conditions, a clear decrease in IASCC was reported, and the decrease occurred even before hardness or RIS were significantly recovered [11,15]. The full recovery of IASCC was only reached when hardening as well as silicon segregation was significantly reduced.

The effects of PIA on the microstructure, as well as the importance of different annealing parameters, in particular the mutual influence of annealing temperature and time on the defect stability in highly irradiated 316 stainless steels, have not yet been studied in detail. Previously, experiments were performed on 304 [15,16] and 316 [12,17] type stainless steels irradiated to moderate doses of 25 dpa. For this paper, the effect of thermal annealing on 316 CW type stainless steel irradiated in a commercial reactor to 80 dpa was studied. Annealing conditions were selected to have a significant influence on the defect structure and to verify if the iron diffusion length is the optimal parameter to describe the defect dissolution.

2. Experimental

The specimens which were used for the annealing experiments were retrieved from an irradiated thimble tube, extracted from the Tihange 2 PWR from position H11 in the reactor core. The tube was irradiated under normal PWR operation conditions. The maximal dose was 80 dpa and the irradiation temperature was around 320 °C [18]. The tube is fabricated from a 316CW type stainless steel and the composition is given in Table 1. A tube segment of 10 mm in length was selected for this study from a part of the tube that was irradiated to 80 dpa. Four equal samples were fabricated and annealed under four different thermal conditions in a furnace installed in a hot cell. The furnace provides the possibility of temperature variation in the range from room temperature up to 1000 °C with a temperature stability in the whole temperature range of the order of 1–2 °C. Annealing was performed under vacuum, with a pressure of about 10^{-5} mBar. The oven was pre-heated to the annealing temperature before inserting the sample. In this way the sample heating time was reduced, ensuring an isothermal anneal.

The annealing parameters are summarized in Table 2. In PIA experiments on stainless steels it is common to combine the effect of the annealing temperature and time in a single parameter, the iron diffusion length. It is assumed that all recovery processes are driven by diffusion processes. The iron diffusion length is calculated using the formula:

$$d = \sqrt{Dt}$$

with t the annealing time (seconds) and D the iron self-diffusion coefficient (m^2/s) given by:

$$D = D_0 \exp\left(-\frac{Q}{kT}\right)$$

where k is the Boltzmann constant, T the annealing temperature (K) and Q the migration energy. Values are taken from Busby et al. [11]: $Q = 1.29$ eV and $D_0 = 1.54 \times 10^{-6}$ m^2/s.

Table 1
Composition of the 316CW stainless steel.

C	Si	Mn	P	S	Cr	Ni	Mo	Co
0.044	0.53	1.79	0.022	0.009	17.40	12.8	2.68	0.07

Table 2
Annealing parameters of the different specimens.

Segment	Anneal T (°C)	Anneal time (h)	Fe diffusion length (cm)
Quarter 1	500	6	0.00114
Quarter 2	550	6	0.00204
Quarter 3	500	19.5	0.00205
Quarter 4	600	6	0.00344

After annealing, a thin slice was cut from the quarter tube and mechanically polished using SiC paper to a thickness of about 0.1 mm. A 1 mm disc was punched out of the polished slice and glued on a platinum 0.6 mm aperture grid with Struers Tripod wax. Finally, the samples were electrochemically polished until perforation by dual jet polishing in an electrolyte consisting of 5% perchloric acid and 95% methanol at −20 °C and applying a voltage of 38 V.

The specimens were investigated in a JEOL 3010 transmission electron microscope operating at 300 kV. Conventional bright field, dark field and weak beam imaging techniques were applied. Frank loops were visualised with the relrod technique and cavities are revealed in under- and over-focused images. The local thickness of the sample was determined with convergent beam electron diffraction (CBED) [19].

3. Results

3.1. As-irradiated material

The defect structure of the thimble tube after irradiation to 80 dpa is shown in the different images of Fig. 1. The typical defects, observed in a stainless steel material irradiated to more than 1 dpa at 300 °C [20], were found in this sample. Namely, the Frank loops (F), black dots and small perfect dislocation loops (SL), are clearly visible in Fig. 1a. The total defect density was found to be $(10.0 \pm 0.5) \times 10^{22}/m^3$.

Fig. 1b is a dark field image obtained by using the relrod diffraction intensity which is induced by the Frank loops. In the inset, the corresponding diffraction pattern is shown. The white circle indicates the part of the diffraction pattern that was selected for the dark field image. It only includes the relrod diffraction intensity. Examination of several relrod images allows the estimation of the total amount of Frank loops and the density was found to be about $(7.2 \pm 0.3) \times 10^{22}/m^3$.

Apart from the Frank loops, round features were observed in Fig. 1b as well, which can be attributed to precipitates. The properties of these precipitates agree with previous observations of γ' precipitates (Ni_3Si) in neutron irradiated 304 and 316 types of stainless steel [21]. Ni_3Si has a fcc crystal structure with a lattice parameter of 0.3505 nm, which is slightly smaller than the lattice parameter of AISI 316 steel. The diffraction patterns are very similar but because the lattice parameter is slightly lower, the locations of the diffraction spots are slightly further away from the transmitted beam than those from the steel. There is no orientation relation with the stainless steel and therefore part of the precipitates generate diffraction spots near the relrods and are visible in the relrod image. Since not all precipitates are visible under the applied imaging conditions, the measured precipitate density of about $0.5 \times 10^{22}/m^3$ is probably an underestimation of the actual density.

Fig. 1c contains an out-of-focus image of the edge of the sample. In this image, cavities appear as white spots and a few examples are indicated with a letter V. The cavities can be either gas filled bubbles or voids. In this paper, we do not differentiate those with or without gas and in the remaining of the text we refer to them as