THE INFLUENCE OF Fe-ION IRRADIATION ON THE MICROSTRUCTURE OF REDUCED ACTIVATION FERRITIC-MARTENSITIC STEEL EUROFER 97

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Abstract

The reduced activation ferritic/martensitic steel Eurofer 97 is the European benchmark structural material for in-vessel components of fusion reactor. Experimental data on neutron irradiated Eurofer 97 have shown a decrease in plasticity and radiation hardening at the irradiation temperatures of ~ 300 °C. The formation of dislocation loops and α’ precipitates is considered as the main reason of these effects. In this work Eurofer 97 was irradiated with Fe ions up to $10^{16}$ cm$^{-2}$ at 250, 300, and 400 °C. The irradiated samples were characterized by TEM and APT. TEM study of ion irradiated samples revealed nucleation of dislocation loops. The pair-correlation analysis of APT data detected an initial stage of solid solution decomposition. The hardening of ion irradiated Eurofer 97 was calculated with DBH model taking into account radiation-induced dislocation loops to compare it with the change of yield stress in neutron irradiated Eurofer 97. According to the obtained results it can be supposed that the formation of dislocation loops plays the main role in the low temperature radiation hardening of Eurofer 97 at the dose level up to ~ 6 dpa.

1. INTRODUCTION

The reduced activation ferritic/martensitic (RAFM) steels are considered as promising structural materials for advanced fusion reactors working in extreme operational conditions at high neutron fluxes ($\sim 10^{13}$–$10^{14}$ n/cm$^2$s) and at elevated temperatures (up to 650 °C). F/m steels show reasonably good thermo-physical and mechanical properties, a low sensitivity to radiation-induced swelling under neutron irradiation and good compatibility with major cooling and breeding materials [1]. The RAFM steel Eurofer 97 is a European benchmark structural material for in-vessel components of fusion reactors, so its extensive studies have been carried out. When irradiated to 2.5, 8.4, and 16.3 dpa in high flux reactor (HFR, Petten) and irradiated to 32 and 70 dpa in BOR 60 (Dimitrovgrad) reactor at temperatures of ~ 300 °C, Eurofer 97 demonstrated degradation of the mechanical properties, i.e., radiation strengthening and therefore the loss of ductility [2, 3]. Detailed transmission electron microscopy (TEM) analysis of its microstructure was carried out in [4, 5]. TEM-visible irradiation microstructural features (voids and dislocation loops formation, MX and M$_{23}$C$_6$ precipitate evolution, and grain boundary segregation) can only partly explain the hardening of irradiated Eurofer 97 [5, 6]. The atom probe tomography (APT) study of Eurofer 97 irradiated in BOR 60 up to 32 dpa at 332 °C revealed nucleation of numerous α’ pre-precipitates [7], which can give additional input to radiation induced strengthening [8].

The objective of the present paper is to provide an analysis of the microscopic origin of degradation of Eurofer 97 under low temperature irradiation. Reactor experiments on radiation resistance of materials are very expensive and time consuming. Nowadays, simulation experiments with heavy ion beams are providing some progress in understanding of radiation degradation of f/m steel. Self-ion irradiation was used in this study of radiation effects in Eurofer 97.

2. MATERIALS AND METHODS

An industrial batch of European RAFM steel Eurofer 97 was produced by Böhler Austria GmbH. To obtain a well-defined martensitic–ferritic structure, Eurofer 97 was subjected to a two-step heat treatment at about
980 °C/30 min, followed by 760 °C/2 h (the heat E83697). This very material was used for the ARBOR and SPICE irradiation programs. The composition of Eurofer 97 steel is presented in table 1.

TABLE 1. THE CHEMICAL COMPOSITION OF EUROFER 97 STEEL (HEAT E83697) UNDER STUDY, WT. %

<table>
<thead>
<tr>
<th>Main elements</th>
<th>C</th>
<th>Si</th>
<th>Mn</th>
<th>Cr</th>
<th>Ni</th>
<th>N</th>
<th>V</th>
<th>W</th>
<th>Ta</th>
</tr>
</thead>
<tbody>
<tr>
<td>Eurofer 97</td>
<td>0.12</td>
<td>0.06</td>
<td>0.47</td>
<td>8.93</td>
<td>0.022</td>
<td>0.018</td>
<td>0.2</td>
<td>1.07</td>
<td>0.14</td>
</tr>
</tbody>
</table>

In our study Fe\(^{2+}\) ion beam accelerated up to 5.6 MeV was used for irradiation. The RFQ HIP-1 output channel was optimized for the experiment, and the target chamber with the sample holder, a heating set and beam control system were installed [9]. According to the SRIM calculations, the maximum number of generated defects in pure Fe under 5.6 MeV Fe\(^{2+}\) irradiation is located at about 1.4÷1.5 μm depth (see Fig. 1).

Disks with a diameter of 3 mm and 0.1 mm thickness were cut out from the bulk material of the Eurofer 97. These disk specimens were mechanically polished up to a thickness of 100 μm. The roughness of the sample surface was measured with atomic-force microscopy before and after irradiation, and it was less than 50 nm for all samples. The disk specimens were irradiated up to \(10^{16} \text{ cm}^{-2}\) at 250, 300 and 400 °C. An estimated dose in the damage peak for this fluence was about 10 dpa.

An analysis of chemical and phase composition was carried out by TEM, electronic diffraction (ED), high resolution TEM (HRTEM), and scanning transmission electron microscopy (STEM). A Titan 80-300 S/TEM (Thermo Fisher Scientific, USA) microscope with the accelerating voltage of 300 kV and equipped with high-angle annular dark field detector (HAADF, Fischione) to provide Z-contrast micrographs was used. The qualitative and quantitative chemical analyses of samples were carried out by the energy dispersive X-ray spectroscopy (EDXS).

Cross sectional samples for S/TEM studies were prepared by the Focused Ion beam (FIB) on a FIB/scanning dual beam microscope Helios NanoLab 600i (Thermo Fisher Scientific, USA). Samples for atom probe tomography (APT) were also lifted out with FIB from the irradiated samples at 0.9-1 μm depth from the irradiated surface. The damage dose at this depth was ~ 6 dpa. APT studies were carried out with APPLE-3D (ITEP) [10] atom probe microscope in the laser evaporation mode with wave length of 515 nm. Data collection was carried out at the sample temperature of 50 K, laser frequency of 25 kHz, and pulse energy of 0.1 - 5 μJ.

**FIG. 1.** The SRIM damage (red solid line) and ion implantation profiles (green dashed line) in pure Fe irradiated with 5.6 MeV Fe\(^{2+}\) ions. The calculations were carried out for a dose of \(1 \times 10^{16} \text{ cm}^{-2}\), using the threshold displacement energy of 40 eV and the Kinchin-Pease option.
3. TEM/STEM MICROSTRUCTURE ANALYSIS

The microstructure analysis was performed on both unirradiated specimens and the ones irradiated with Fe\(^{2+}\) ions up to \(10^{16}\) cm\(^{-2}\) at 250, 300 and 400 °C. For the quantification of radiation induced defects, bright field (BF) and dark field (DF) images were acquired. Micrographs for estimation of the dislocation loops density and diameters were acquired from the grains with orientation close to the [111] zone axis. In addition, the dislocation loops were highlighted using weak beam dark field (WBDF) imaging in (g, 3g) diffraction conditions.

Fig. 2 shows the microstructure of the Eurofer 97 sample after irradiation with Fe\(^{2+}\) ions at 300 °C. At no irradiation temperatures used in the experiment, any significant microstructural changes (prior austenite grain size, morphology, size and distribution of the second phase precipitates) were observed by TEM. The microstructure of the Eurofer 97 steel in the as-received condition consisted of tempered martensite with a prior austenite grain size in the range between 6 and 11 µm. Small fraction of spherical particles with the size of 30-90 nm enriched in W and Ta were observed inside the grain structure. Also large disk shaped carbides were detected at the boundaries between martensite plates.

Radiation induced defect clusters and dislocation loops were observed in all irradiated specimens. On the micrographs they had a contrast as a black dots or typical “coffee bean” pattern that could be highlighted using WBDF method (Fig. 3). The calculated number density of defects at various depths correlated with the radiation damage profile (Fig. 4). To estimate the thickness of the areas chosen for quantification, the converged beam diffraction technique was used. An analysis of the radiation induced changes was carried out at the depth of ~800-1000 nm, where damage dose was ~ 5-6 dpa. The calculations showed that with the increase in irradiation temperature, the density of dislocation loops reduced. The loop number density was estimated \(2.4 \times 10^{22} \text{ m}^{-3}\) at 250 °C, \(1.7 \times 10^{21} \text{ m}^{-3}\) at 300 °C and \(7.7 \times 10^{21} \text{ m}^{-3}\) at 400 °C. The loop diameter increased with increasing the irradiation temperature (Fig. 5). The calculated average diameter of the loops in the specimens irradiated at 250, 300 and 400 °C was 8, 10, and 12 nm correspondingly.

![Fig. 2. STEM BF image of the microstructure of Eurofer 97 irradiated with 5.6 MeV Fe\(^{2+}\) ions to \(10^{16}\) cm\(^{-2}\) at 300 °C and a damage dose profile.](image)
4. ATOM PROBE TOMOGRAPHY

The comparison of the atom maps obtained for un-irradiated and ion-irradiated specimens did not show any signs of visible precipitation or clustering processes. In some volumes large features like grain boundaries or carbides (Fig. 6) were detected. The homogeneity of the atoms spatial distribution was tested using pair correlation function analysis [11]. The correlation analysis was completed in $15 \times 15 \times 30 \text{ nm}^3$ volumes extracted away from the large features. A slight increase (~ 5%) in the pair Cr-Cr correlation function was observed at small distances (< 4 nm) in the specimens irradiated at 400 °C (see Fig. 7). This effect corresponds to the nucleation of chromium enriched embryos with the sizes of about 4 nm. The detected increase in the pair correlation function is considerably lower than the one detected in Eurofer 97 irradiated with neutrons at 330 °C to 32 dpa [7, 8]. It allows to suggest that in the steel irradiated with heavy ions up to 6 dpa, chromium enriched embryos make only a small input to hardening.

![Fig. 3. BF (a) and (g, 3g) WBDF (b) images showing radiation induced defects in Eurofer 97 irradiated with 5.6 Fe$^{2+}$ ions to 6 dpa at 300 °C.](image)

![Fig. 4. Calculated SRIM damage profile (line) and loops number density (points) at different irradiation temperatures.](image)
FIG. 5 Dislocation loops size distribution at a depth of 850 nm depth.

FIG. 6. The atom maps showing tungsten carbide in Eurofer 97 irradiated up to 6 dpa at 400 °C.

FIG. 7. The correlation Cr-Cr function of Eurofer 97 irradiated with Fe ions up to 6 dpa.
5. RADIATION INDUCED HARDENING

The contributions to hardening from different short range obstacle types in the framework of the Dispersed Barrier Hardening (DBH) model are in general considered by a root sum square rule [12] as described by

$$\Delta \sigma_{\text{tot}} = \sqrt{\sum \Delta \sigma_i^2}, \quad \text{where} \quad \Delta \sigma_i^2 \quad \text{is the change due to the } i\text{-th obstacles such as loops, voids, precipitates, etc.}$$

The hardening $\Delta \sigma_i$ caused by the interaction of moving dislocations with obstacles of type $i$ is

$$\Delta \sigma_i = M_T \alpha_i \mu b \sqrt{N_i d_i},$$

where $\mu$ is the shear modulus of the matrix, $b$ is the Burgers vector of gliding dislocations, $\alpha_i$ is the barrier strength, $M_T$ is the Taylor factor, and $N_i$ and $d_i$ are the number density and the mean size of obstacles respectively.

The contribution to hardening from dislocation loops of $\Delta \sigma_{\text{loops}}$ was calculated in the framework of the DBH model with the values of the barrier strength of $\alpha_{\text{loops}} = 0.25$, and 0.4, and $\mu = 84.4$ GPa, $M_T = 3.06$, $b = 0.248$ nm (see Fig. 8). The hardening calculated with $\alpha_{\text{loops}} = 0.4$ [13] correlates with the increase of yield strength of neutron irradiated Eurofer 97 [14].

![Dislocation loops hardening vs depth for Eurofer 97 for different barrier strength coefficients $\alpha$.](image)

FIG. 8. Dislocation loops hardening vs depth for Eurofer 97 for different barrier strength coefficients $\alpha$.

The comparison of neutron and ion irradiation experimental results involves a lot of effects that should be taken into consideration [15]. We have compared the hardening calculated with DBH model, which takes into account radiation-induced dislocation loops (see Fig. 8), with the change of yield stress for neutron irradiated Eurofer 97 acquired in the SPICE irradiation project [16, 17]. In our calculations the value of the barrier strength of $\alpha_{\text{loops}}$ was taken 0.4, which provided a good agreement with the yield strength of neutron irradiated Eurofer 97 [14]. The temperature shift calculated according to Mansur [18] was used to take into account the dose rate effect in the ion irradiation experiment. The comparison of hardening values obtained in the ion and neutron irradiation experiments (see Fig. 9) has showed a reasonable agreement. It seems that the formation of dislocation loops plays the main role in the low temperature radiation hardening of Eurofer 97 at the dose up to ~ 6 dpa.

6. RESULTS

In this work, the analysis of the microscopic origin of degradation of Eurofer 97 under low temperature irradiation with Fe ions to $1 \times 10^{16}$ cm$^{-2}$ at 250, 300 and 400 °C was carried out. The irradiated samples were characterized by TEM and APT. A preferable formation of dislocation loops was detected after the irradiation at all the temperatures used in the study. The pair-correlation analysis detected an initial stage of solid solution decomposition in Eurofer 97 irradiated to 6 dpa at 400 °C. The detected microstructural changes and the hardening calculated with DBH model conform to the results for the neutron irradiated Eurofer 97.
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