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Transmission electron analysis of dislocation loops in T91 steels from MEGAPIE and MIRE irradiation experiments

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ABSTRACT

T91 steel samples were irradiated in both a neutron and a neutron plus proton irradiation environments, and the resulting microstructure was studied using transmission electron microscopy. Three samples were irradiated with neutrons and protons to a cumulative dose of 0.88, 2.07, and 4.35 dpa at temperatures of 254 °C, 260 °C and 320–350 °C, respectively and two samples were irradiated with neutrons to 0.6 and 1 dpa, at a temperature of 290 °C for comparison. Radiation induced dislocation loops were observed in both groups of samples. In all samples irradiated up to 1 dpa, irrespective of irradiation conditions, dislocation loops were segregated near the line dislocations, forming a clear heterogeneous distribution. In the high dose sample, on the other hand, they were uniformly distributed over the grain interior and the middle dose sample showed a mixed character. Voids of similar average size were only observed in the middle and high dose samples, and their appearance seems to be correlated with the onset of homogeneous loop distribution. The observed defect properties are rationalized on the basis of modeling results.

1. Introduction

High Cr ferritic/martensitic steels are candidate structural materials for key components in several Generation IV concepts, accelerator driven systems (ADS) and fusion reactors because they are highly resistant to neutron irradiation and provide good resistance to the corrosion [1–3]. These reactors will operate under challenging conditions such as high operating temperature (e.g. above 450°) and neutron fluence. In addition, new potentially aggressive coolants are foreseen to be used. For example, in an ADS like MYRRHA the structural materials need to withstand temperatures up to about 550 °C, neutron damages of the order of several dpa, while being in contact with lead bismuth eutectic which is both coolant and spallation target material [4]. These conditions can substantially enhance the material degradation by causing hardening, embrittlement, swelling, creep, fatigue, etc. [5–9]. Therefore, the structural integrity of the materials after irradiation needs to be fully evaluated. This includes the study of both the mechanical properties and microstructural evolution under irradiation. A variety of degradation effects, in particular liquid metal embrittlement (LME) [10] might affect the applicability of certain materials in the MYRRHA reactor system. In spite of large efforts, this effect which is characterized by significant reduction of total elongation in a tensile test experiment, is still not fully understood. One of the open issues is related to the lack of physical understanding of the synergetic effects between neutron and proton irradiation to the evolution of the defects in the material.

Among the high Cr ferritic/martensitic steels, the T91 exhibits high strength at elevated temperatures, excellent creep behavior, and resistance to thermal fatigue [11]. In a previous study, the mechanical properties of T91 steel which was exposed to simultaneous proton/neutron irradiation and in contact with liquid metal environment were analyzed [12]. Two distinctly different conditions were compared. In the first one, provided by the Megawatt pilot experiment (MEGAPIE), the T91 steel is exposed to a proton/spallation neutron irradiation in combination with exposure to a flowing liquid lead-bismuth eutectic (LBE). In the second condition, provided by the TWIN-ASTIR experiment, the T91 steel is exposed to neutron irradiation under stagnant lead-bismuth conditions. Under both conditions, a significant influence of liquid metal on the tensile properties, and in particular the reduction of total elongation was observed. The reduction of total elongation increased linearly with increasing dose. The different conditions in two irradiation programs, such as irradiation temperature

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fluctuations, the presence of neutron/proton irradiation, with and without the contact with LBE, were found to induce no significant differences in the mechanical properties of irradiated T91.

In this study, the microstructure of three T91 samples irradiated in the MEGAPIE experiment, and two T91 samples irradiated in the material irradiation experiment (MIRE) were studied with transmission electron microscopy (TEM). The formation of radiation induced defects and their evolution as a function of dose were analyzed and compared with similar results in other high Cr alloys and steels. The observed defect properties and behavior are rationalized on the basis of modeling results.

2. MEGAPIE and MIRE irradiation experiments

MEGAPIE was a joint world-wide initiative to design, build, operate and explore a liquid lead-bismuth spallation target for 1 MW of beam power [13,14]. With respect to material issues, the goal of this experiment is to acquire relevant materials data for a design database for liquid metal targets. By taking advantage of the spallation facility (SINQ) at Paul Scherrer Institute (PSI), a target was developed and operated for about four months. The T91 material used in this study is extracted from the proton beam window area, the so-called calotte of the LBE container. The fluence and the temperature distributions in the beam window T91 material, together with the tensile sample extraction scheme are presented in Refs. [15,16]. The tensile samples which are tested in this study received a radiation dose (protons and neutrons) of 0.8–3.9 displacements per atom (dpa) under large temperature fluctuations due to frequent beam trips of the proton beam. The temperature variations were in the range of 250–350 °C depending on the position in the calotte [15,16]. By the end of the irradiation, the MEGAPIE target has received about 3 Ah proton charge, which corresponds at the beam window to a peak proton fluence of about $2 \times 10^{25} p/m^2$ and a maximum irradiation dose of the order of 8 dpa. The irradiation conditions of the samples which were cut from the calotte and sent to SCK•CEN from PSI are summarized in Table 1.

In the MIRE experiment, neutron irradiation was performed in the BR2 material testing reactor of SCK•CEN. The irradiation conditions are presented in Table 1. The cut-off energy of the neutron spectrum in the BR2 reactor is about 8.5 MeV. The neutron flux was around 7.4×10^{13} n/cm^2/s. Three fluences were reached, corresponding to one, three and five cycles of the reactor. Taking into account the results of the dosimetry, that depend on the power of the reactor and the neutron spectrum, these fluences are calculated to correspond to 0.06, 0.6 and 1.0 dpa [17]. In this study the focus is given to middle (0.6 dpa) and high (1.0 dpa) fluence levels since they are comparable to the low dose MEGAPIE sample.

3. Materials and testing

The material composition of the irradiated T91 steels is given in Table 2. The details about the preparation of MEGAPIE irradiation experiments and the sample extraction can be found in Refs. [15,16,18]. The MIRE irradiation program is described in Refs. [17,19].

The samples were mechanically polished in a hot-cell on SiC paper to reduce the thickness of the discs to about 0.1 mm. Afterwards, they were electrochemically polished in fumehood until perforation. The electrolyte that was used consisted of 5% perchloric acid in 95% methanol and they were polished at a temperature of -20^0C applying a voltage of 20 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. The local thickness of the sample was determined with convergent beam electron diffraction (CBED). The dislocation density (ρ) was measured by counting all dislocation lines (N) intersecting a circle of diameter d as $\rho = 2N/\pi dt$ [20], where t is the local sample thickness. A correction factor is applied for extinction conditions assuming that all dislocations are $a_0/2\langle 111\rangle$ type. The number density of the small defects was calculated by counting the number of visible defects in a TEM image and dividing by the sample volume. A correction factor is applied to account for defects that are extinct due to the invisibility criterion $g \cdot b = 0$. For dislocation loops it is assumed that all loops have an $a_0\langle 100\rangle$ type burgers vector, as will be demonstrated from extinction conditions, and hence for image recorded with a $\mathbf{g} = 200$ type diffraction vector, the number of visible defects is multiplied with a factor of 3, while for a $\mathbf{g} = 110$ type diffraction vector, the number is multiplied with a factor 3/2. All dislocation and defect densities were determined at 3 different areas, at least, and the average density is calculated. The size of the dislocation loops was measured along the long axis in either bright field or weak beam images. It should be considered that the defect contrast does not match the exact size of the defect. Under some conditions the contrast is on the outside of the loop, while under opposite conditions, they are on the inside. Defects were selected from different areas and under different conditions, such that the deviations from the exact size cancel out when calculating the defect density. The number of loops that were measured is given when the average value is mentioned. The voids are revealed in under and over focus images with a defocus value around 1–1.5 μm. One has to keep in mind that neutron irradiation could produce some He. Since the electron energy loss spectra were not completed, neither could faceting be seen with such small defect sizes (to confirm that it is indeed a void), it would be more appropriate to classify these defects as cavities. However, since the concentration of He is expected to be low, a void is used as a nomenclature for these defects in the rest of the text. The size of the voids was measured between the edges of the Fresnel contrast and the average value is calculated.

4. Transmission electron microscopy

The TEM micrographs of non-irradiated T91 steel are shown in Fig. 1. The T91 material has a ferritic/martensitic grain structure. Carbides are formed preferentially at the grain boundaries. A high dislocation density was observed, of about $\pm 1 \times 10^{13}/m^2$ which is typical for the ferritic/martensitic structure.

Table 1
MEGAPIE and MIRE irradiation conditions. Complex flux distribution in the MEGAPIE window is provided in Refs. [15,16,18].

Experiment	material	Dose (dpa)	Temperature (°C)	flux (n/cm^2/s)	Irradiation
MIRE	T91	0.6	290	7.4×10^{13}	neutrons
	T91	1.0	290	7.4×10^{13}	neutrons
MEGAPIE	T91	0.88	254		neutrons + protons
	T91	2.07	260		neutrons + protons
	T91	4.35	320–350		neutrons + protons

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