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TEM study and modeling of bubble formation in dual-beam He^{+}/Fe^{3+} ion irradiated EUROFER97

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HIGHLIGHTS

- Investigation of He and dpa effects in EUROFER97 by irradiation with He/Fe-ions at 330 °C, 400 °C and 500 °C.
- TEM analysis of the size distribution of helium bubbles as a function of irradiation temperature.
- Modeling of helium bubble formation with a kinetic rate model.
- Influence of two different thermodynamic descriptions of helium bubbles on the rate model.
- Comparison between experimental and numerical results.

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ABSTRACT

The Reduced Activation Ferritic/Martensitic (RAFM) steels are promising structural materials for the first wall and blanket components of future fusion reactors. To obtain further insight into the temperature dependence of helium effects induced by 14 MeV energy neutrons under fusion like conditions, EUROFER97 was exposed to He^{+}/Fe^{3+} dual-beam ion irradiation at the JANNUS laboratory at Saclay. The implantation was carried out at temperatures of 330 °C, 400 °C and 500 °C and induced a damage and helium concentration up to 26 dpa and 450 appm He, respectively. TEM microstructure analysis indicates a spatially homogeneous distribution of helium bubbles at 330 °C and 400 °C whereas a coexistence of homogeneous and heterogeneous nucleation of bubbles is found at 500 °C. An increasing mean bubble diameter and decreasing concentration of bubbles with rising irradiation temperature, as predicted by numerical results of a kinetic rate model for diffusion governed homogeneous nucleation of helium bubbles, are mostly confirmed by the irradiation experiment. Furthermore, within the rate model two approaches for the determination of the thermodynamic properties of helium filled voids in α-iron are applied. With respect to the final bubble size distribution, the commonly used surface energy of a void in the iron matrix is compared to the "variable gap model" of [1], J. Nucl. Mater. 418 (2011), which includes additionally the interaction between the helium atoms themselves, the energy at the helium-iron interface and the elastic deformation of the iron matrix.

1. Introduction

The reduced activation ferritic-martensitic steels are considered as potential structural materials for the first wall and blanket components of future fusion reactors due to their promising low irradiation induced swelling and well balanced physical and thermo-mechanical properties [2]. Nevertheless, when exposed for several years to a high flux of 14 MeV neutrons in fusion reactors these materials are still expected to sustain severe damage. Accordingly, a detailed knowledge of the evolution of microstructure damage is crucial for the estimation of the mechanical degradation of structural components and the further development of accurate design limits.

Due to the absence of facilities with fusion reactor relevant neutron spectra substitutional irradiation programs are executed to simulate the mechanisms of void nucleation under simultaneous

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production of atomic displacement cascades and helium. Aside from the here discussed dual-beam irradiation with iron and helium ions, helium effects in EUROFER97 were studied in the fission neutron irradiation programs SPICE and ARBOR by using boron doping technique [3–6] and in spallation proton irradiation programs STIP-II and STIP-III [7,8] including also protons and helium ions in addition to neutrons.

For the modeling of helium effects in α-iron a couple of theoretical methods is applied, providing insight on different time and length scales. On atomic scales density functional theory [9] and molecular dynamics [10,11] provide insight into properties of interstitial helium and small helium-vacancy clusters embedded in an iron-matrix, like migration and binding energies, and support the transfer to macroscopic thermodynamic description of helium bubbles [1,12]. Based on the knowledge of these physical quantities, usually object kinetic Monte Carlo calculations, kinetic rate theory or the Fokker-Planck equation are used to trace the evolution of microstructural damage on times and length scales typical for radiation experiments [4,13–16].

At the moment, the damage of 14 MeV neutrons as well as its reasonable imitation are not sufficiently understood and of great importance for the development and verification of suitable materials. Particularly, it was found that helium in EUROFER97 is responsible for embrittlement [17] and for an increase of the nanohardness [18]. Accordingly, the temperature dependence of helium bubble formation induced by He^{+}/Fe^{3+} dual-beam ion irradiation of EUROFER97 is analyzed with TEM investigations of the irradiated samples. Beside the experimental part, the understanding of helium effects is supported by a kinetic rate model for diffusion governed homogeneous nucleation of helium bubbles. In this manner, we will evaluate the qualitative and quantitative capabilities of the model for nucleation of helium bubbles in EUROFER97 in order to assess the need for extending the model or adjusting parameters.

2. Dual-beam ion irradiation and sample preparation

The 25 mm thick EUROFER97 plates were produced by Böhler Austria GmbH. The final heat treatment applied by the manufacturer includes a normalization at 980 °C for 0.5 h and tempering at 760 °C for 1.5 h. For the ion irradiation discs of 3 mm diameter and approximately 90 μm thickness were prepared. The irradiation with 1.2 MeV He^{+} and 3.0 MeV Fe^{3+} ions was performed at the JANNUS Facility at Saclay [19,20] at temperatures of 330° C, 400 °C and 500 °C. We achieved a broadened implantation profile of helium by inserting aluminum energy degraders into the beam. In order to choose ion doses which lead to damage and helium

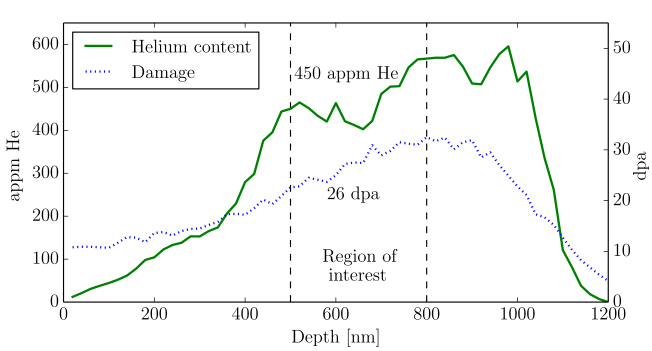

Fig. 1. SRIM calculation of the damage and helium concentration profile achieved by 6 h of 1.2 MeV He^{+} and 3.0 MeV Fe^{3+} ion irradiation with respective doses of $3.2\cdot10^{16}$ Fe^{3+} cm^{-2} and $1.1\cdot10^{16}$ He^{+} cm^{-2}. The desired damage and helium content of approximately 26 dpa and 450 appm He is achieved at implantation depths in between 500 nm and 800 nm.

concentration ratios similar to conditions expected in future fusion reactors, SRIM [21] calculations were performed. Fig. 1 displays the expected depth profiles of damage and helium concentration for the applied doses of $3.2\cdot10^{16}$ Fe^{3+} cm^{-2} and $1.1\cdot10^{16}$ He^{+} cm^{-2}. Thus, fusion like conditions are obtained at depths between 500 nm and 800 nm from the surface where the applied fluxes lead to 26 dpa/450 appm He and a helium-dpa ratio of approximately 15 appm He/dpa.

TEM samples were prepared via electrolytic-polishing or focused-ion-beam (FIB) machining. In particular, attention has to be paid to the electrolytic-polishing procedure for the creation of a thin film at a depth with the desired damage and helium content. Accordingly, the disks were thinned separately from the front and back side in an asymmetric manner. A more detailed description of the specimen preparation, the irradiation experiment and the TEM sample preparation can be found in Ref. [22]. The preparation of TEM-samples out of the uppermost irradiated 1 μm thick layer with asymmetric electrolytic-polishing and FIB machining may fail due to thinning at the wrong depth and the additional ion irradiation, respectively. In order to avoid features in the microstructure introduced by the sample preparation for all irradiation temperatures samples were produced in both ways. The observation of the same qualitative results in case of both preparation methods therefore ensures that they do not dependent on the preparation method. Nevertheless, for the subsequent detailed quantitative evaluation of size and density of helium bubbles we used FIB samples to guarantee for the results depths of 500 nm–800 nm. Thus, we assure a damage and helium content of 26 dpa and 450 appm He, respectively.

3. TEM-analysis of the helium bubble distribution

The irradiation damage is analyzed via TEM bright-field technique to obtain size distributions of voids. As well accepted, voids are identified by changing of their contrast from a bright spot with a dark fresnel fringe in under-focus condition to a dark spot with a bright fresnel fringe in over-focus condition. Under the assumption of spherical voids, their size is determined by measuring the corresponding diameter in an under-focused micrograph. The subsequent results and micrographs stem all from samples prepared with FIB machining and refer to the region of interest at a depth in between 500 nm and 800 nm, as marked in Fig. 1. Besides cavities other defects of the microstructure as dislocation loops and precipitates characterize the irradiation damage. The in depth investigation of these types of defects will be subject of future publications. With respect to the application of rate-model we limit the present TEM study to the size and density of helium bubbles. For characteristics of dislocations necessary for modeling we will refer on existing studies on EUROFER97 for dual-beam irradiation [22] and neutron irradiation [5,23,24].

In the case of $T_{irr} = 330$ °C we find homogeneously distributed bubbles as displayed in the two micrographs of Fig. 2. Similar observations are obtained for specimens irradiated at a temperature of $T_{irr} = 400$ °C, shown in Fig. 3. The bubbles are distributed homogeneously in the matrix without large variations in size. Remarkably, rising the irradiation temperature from 330 °C to 400 °C does not qualitatively influence the formation of helium bubbles. In contrast, further increasing the irradiation temperature up to $T_{irr} = 500$ °C strongly influences the nucleation of bubbles, concerning their size as well as spatial distribution. In contrast to the low temperature irradiation a heterogeneous nucleation is observed at $T_{irr} = 500$ °C. A major part of the bubbles is still homogeneously distributed in the matrix of the steel, as shown in Fig. 4. In addition, a remarkable fraction of the bubbles are now nucleated at different extended microstructural sinks such as grain