Correlation of microstructure and nano-mechanical property in ion-irradiated Fe based alloys

Chuanxin LIU
Candidate for the Degree of Doctor
Supervisor: Somei Ohnuki
Division of Materials Science and Engineering

Introduction

Ferritic/martensitic steels, which have excellent irradiation damage resistance, are candidates for fusion reactors. However, irradiation causes undesirable material degradation such as irradiation hardening, which may make their use impossible at low temperatures (<0.4T_m, >0.1 dpa). Irradiation hardening is believed to be due to defect clusters, dislocation loops or lines, voids and bubbles, and other changes, which are produced by energetic neutron-atom collisions and nuclear transmutation reactions. To simulate high energy neutron irradiation, high energy self-ions have been employed. This study focused on that the correlation of microstructure and nano-mechanical property in ion-irradiated Fe based alloys.

Experimental procedure

In current work, Fe-8Cr model alloy and F82H-ODS steel were mainly selected target materials. In comparison, pure iron, F82H (ferritic/martensitic steel), and two classes of ODS steel (316-ODS and High Cr-ODS steels) were also investigated. Fe-8Cr model alloy is a predominant material of particular interest for irradiation damage studies since it is a major constitute in many ferrous structural alloys, including the ferritic/martensitic steels which are already considered as candidates for future fast-reactor core components and the first wall of fusion reactors.

In this work, high energy ion irradiation was carried out at TIARA (Takasaki Ion Accelerators for Advanced Radiation Application) facility in JAEA (Japan Atomic Energy Agency). As it shown in Fig.1, this irradiation facility has three branches which response to 3 MV Tandem accelerator, 0.4 MV ion implanter and 3 MV single-ended accelerator. Single ion irradiation with 10.5 MeV Fe^{3+} ions and dual ion irradiation with both 10.5 MeV Fe^{3+} ions and 1.05 MeV He^{+} ions were employed. It is conventional believed that Fe ions, which are the self-ions of steels, were selected to simulate the damage induced by fast neutrons for several reasons including avoidance of activated material due to neutron irradiation, reduction of irradiation times and improved capabilities to vary irradiation conditions. In addition, Fe ions can be expected to avoid the second phase formation owing to the change of chemical composition at local region.

According to SRIM 2008 simulation, Fig.2 plots the damage profile generated for 10.5 MeV Fe^{3+} ions as an
Fig. 2 Depth profile of displacement damage in Fe-8Cr model alloy calculated with SRIM code.

Post-irradiation test was carried out using the Elionix ENT-1100a (Elionix Inc., Japan) nano-indenter with Berkovich diamond indenter tip. Indentation test was carried out at room temperature with 400 nm depth control and the direction of indentation was chosen to be in parallel to the ion beam axis, normal to the irradiated surface. The nano-indentation results were analyzed in manner described by Oliver and Pharr.

A Hitachi FB-2100 Focused Ion Beam (FIB), operated at 40 kV with the Ga⁺ ion source, was used for the preparation of the cross-section TEM foils of the irradiated samples. Furthermore, FIB-ed thin foils were sliced again using a Low Energy Gun with the energy lower than 1 kV down to 15 V in order to remove gallium effect which was introduced during FIB fabrication. TEM observation was carried out using a JEOL JEM-2000FX microscope operated at 200 kV. The local foil thickness was measured using thickness fringes in weak beam dark field images.

Results and discussions

- **Irradiation hardening and microstructures in Fe-8Cr model alloy**

In general the irradiation hardening can be defined by the difference in the hardness before and after irradiation, \( \Delta H = H_{\text{irrad.}} - H_{\text{unirrad.}} \). Log-log plots of irradiation hardening versus displacement dose level are illustrated in Fig.3. Hardness measurement in each irradiation condition for both un-irradiated and irradiated regions was the average of at least 10 available indents to assure reproducibility and reliability.

Firstly, irradiation-hardening versus dose curves showed hardening in this alloy among all irradiated conditions! Secondly, it should be pointed out that over the same dose range irradiation at lower temperature tends to exhibit larger irradiation hardening. Moreover, the dose dependence showed good agreement with a simple power law expression, \( \Delta H \approx (\text{dpa})^n \), where dpa is the irradiation dose, which can be replaced by a factor related to defect cluster barriers, or by the radiation fluence. It is obvious that there exists a reduction in the slopes of log-log plots with the decrease of irradiation temperature. As inserted in Fig.3, the values for the exponent n, representing the slopes of the log-log plots, changed from 0.26, 0.17 to 0.09 with the change of temperature from 300, 250 to 100 °C, respectively. The decrease of exponent n values with decreased
temperature is probably due to a tendency towards saturation region in irradiation hardening at lower irradiation temperature, usually because of irradiation induced cascade overlap.

![Log-log plots of irradiation hardening versus displacement dose in several temperatures](image)

**Fig. 3** Log-log plots of irradiation hardening versus displacement dose in several temperatures

The defect of microstructure in irradiated specimens was investigated in cross-sectional foil using conventional TEM techniques. **Fig. 4** summarized the typical cross-sectional bright field microstructures for Fe-8Cr irradiated to doses of 0.1-10 dpa with single ion irradiation and another dual ion beam irradiation (10 dpa Fe\(^{3+}\) and simultaneous 200 appm He\(^+\) per dpa) at 300 °C.

In present work, displacement dose level was set that a designated dose at a depth of 1 µm from irradiation surface.

![Bright field image of Fe-8Cr irradiated](image)

**Fig. 4.** The bright field image of Fe-8Cr irradiated to doses of (A) 0.1, (B) 1, (C) 10 dpa with single ions, and (D) dual beam irradiation at 300 °C.

TEM examination revealed that there is no apparent visible defect clusters at 0.1 dpa condition; meanwhile, very weak defect clusters were detectable at 1 dpa condition. For this reason, there is no available data for 0.1 and 1 dpa evaluation. In order to understand the origin of the radiation hardening in Fe-8Cr model alloy, the dispersed obstacle hardening model is used as a conventional theory. The measured and calculated hardening due to irradiation-induced defects was investigated. Radiation hardening due to these micro-structural features was calculated by using the dispersed barrier hardening model: \( \Delta \sigma = M \alpha \mu b (N d)^{0.5} \), where M, \( \alpha \), \( \mu \), b, N and d are the Taylor factor (M=3.06), barrier strength of obstacles, the shear modulus of the matrix, the magnitude of burgers vector of moving dislocation, the number density of obstacles and the
mean diameter of obstacles, respectively. The hardening was estimated by using the relation between yield stress change and nano-hardness change derived from non-irradiated alloy and following the parameters: \( \alpha = 0.3 \) for loops. The comparison between calculated hardening and measured hardening indicated that visible defect clusters contributed hardening does not agree with the measured hardening.

As far as is known, the microstructure evolution as a consequence of the formation, migration, agglomeration, and annihilation of point defects, the irradiation condition, for example, temperature, dose and dose rate, should have a large effect on the final size, density and distribution of the defect structures. As mentioned in Fig.4, there was no visible defect clusters with a dose of 0.1 dpa irradiation, however, good developed loop rafts structure were observed in 10 dpa irradiation condition (as shown in Fig.4c). It would not be unreasonable to speculate that the displacement dose level was the main factor for these different micro-structure observation results in the present target materials. On the other hand, quantitative hardness measurements indicated that the non-negligible irradiation-induced hardening was also detected in the lowest irradiation condition (0.1 dpa). This irradiation-induced hardening should be attributed to the remnant defects and/or defect clusters yielded in displacement cascades. In this case, it suggested that there has a threshold dose below which defect clusters cannot be observed by current transmission electron microscopes. Furthermore, it also suggested that the threshold dose for defect clusters visibility was near 0.1 dpa in ion irradiated Fe-8Cr. This is, however, different and almost two orders of magnitude higher than neutron irradiation result (~0.001 dpa was generally mentioned as threshold dose in published papers) for defect clusters visibility in pure iron and ferritic model alloy, even though the difference of the threshold dose between the alloys or steel is explained by the trapping effect of the point defects with impurities. In generally, it is suggested that quantification of small defect clusters can be best evaluated by using weak beam electron microscopy. S.J.Zinkle et al suggested that the observation of small defect clusters near TEM resolution limit must be performed in very thin (< 40 nm) foil region in order to avoid their poor contrast in thicker region with surrounding matrix. According to S.J.Zinkle’s suggestion, the thickness effect might be possible reason of observation dimension limitation for visible small defect clusters in each condition in current work. Furthermore, these small defect clusters contributed to irradiation hardening can diminish the discrepancy between measured and calculated hardening, and it also decrease the threshold dose for defect clusters visibility.

- **Irradiation hardening and microstructures in F82H-ODS steel**

Fig.5 shows irradiation-hardening at different temperatures in F82H-ODS steel irradiated to dose of 20 dpa with single Fe\(^{3+}\) ions. Firstly, irradiation-hardening was detected. Secondly, for two constant temperature conditions, there has a hardening decrease from 1.4 to 1.1 GPa with a temperature increase from 250 to 380 °C. On the other hand, the variable temperature showed the lowest irradiation hardening (0.2 GPa) after the same irradiation dose. Nano-hardness results suggested that irradiation hardening not only depends on the displacement dose, but also can be strongly affected by the irradiation temperature.

![Hardening vs Temperature](image_url)

Fig.5 Irradiation-hardening at different temperatures in F82H-ODS steel with single Fe\(^{3+}\) ions to 20 dpa.

Fig.6 showed the typical bright field image of
F82H-ODS steel with low magnification. The transformed scale in the depth range from the beam incident direction is described as follows. Dislocation lines and precipitates such as carbides (mainly Cr$_2$C$_6$) can be clearly observed in the whole image. Small black dots were observed in a range from incident surface to the peak damage area which response to the depth of approximate 2.5 μm. Moreover, the distribution of black dots in irradiated region is also agreement with SRIM code simulation. Due to no black dots were observed over this depth range, it can be confirmed that those defects should be induced during ion irradiation. Fig.6 (b)-(d) showed the enlarged details of the areas within the rectangle marked in Fig.6(a).
It is well known that the irradiation temperature can have a significant influence on the microstructural evolution in materials. Weak beam dark field images of irradiation induced dislocation loops in three temperature conditions taken with $g = (110)$, $S_3 g > 0$, $g/3g$ close to the [001] zone in the foils at a depth of around 1 μm are shown in Fig. 7. Firstly, it can be found that irradiation induced defects are mainly dislocation loops in small size, which are visible in the weak beam dark field images as white dots. In the case of 250 °C, as it shown in Fig. 7(A), defect clusters showed almost homogeneous distribution over the entire specimen. Small defect clusters with the size between 4 and 6 nm in diameter were mainly formed in this temperature condition. Meanwhile, a few larger defect clusters with a size of about 10 nm can be observed as well. In the case of 380 °C, as it shown in Fig. 7(B), defect clusters can be observed in whole area, there has an increase in clusters size. Larger clusters which indicated the coalescence and rearrangement of small defect clusters or point defects were observed obviously. For the variable temperature irradiation at 300/450 °C, as it shown in Fig. 7(C), the defect clusters had homogeneously distribution. And the size of clusters between 4 and 6 nm in diameter was predominant. Furthermore, smaller defect clusters with the size of 2 nm were observed as well. It suggested that for the constant displacement dose level, microstructure evolution is affected by irradiation temperature. With the change of irradiation temperature, the mobility and agglomeration of small size clusters are changed.
During the comparison of the irradiations with two constant temperatures, a simple consideration of the nucleation of point defect clusters at low temperature and their growth at higher temperature was sufficient to understand the phenomena in current material. Kiritani elucidated that the temperature dependence of defect structure development mechanism. In this case, the interstitial type dislocation loops nucleated by the initial lower temperature irradiation grow rapidly during the higher temperature irradiation and might lead to a complex dislocation structure, whereas no dislocation structure other than that developed from pre-existing dislocations will appear when the irradiation occurs at the higher temperature from the beginning. On the other hand, an important consequence of the defect reaction is considered as the primary candidates of blanket structural materials for fusion reactors, because of the most matured technology base and good resistance to neutron irradiation. Fe-8Cr based alloys are the reference alloys of commercial ferritic/martensitic steels developed around the world. The qualification of these RAFM steels for practical application requires an exhaustive understanding of their microstructure and mechanical the lowest number density and smaller mean size with the variable temperature irradiation were detected. It suggested that the accumulation of more defects at lower temperature leads to fewer defects in the final state because of the more efficient annihilation by the temperature change. Nucleation of interstitial loops was suppressed remarkably by changing irradiation temperature periodically. They suggested that this phenomenon is not thought to be the result of thermal annealing but to be caused by reaction of point defects and their clusters formed at both lower and higher temperature.

In order to understand the origin of the irradiation hardening in F82H-ODS steel, the dispersed obstacle hardening model is used as a conventional theory. The measured and calculated hardening due to irradiation-induced defect clusters was investigated. Radiation hardening due to these micro-structural features was estimated by using the dispersed barrier hardening model: \( \Delta \sigma = M_\alpha \mu_\beta (N_d)^{0.5} \), the same as that mentioned before. It can be deduced that calculated hardening resulting from this suggested model were 0.72, 0.35 and 0.19 GPa for 250, 380 and 300/450 °C, respectively. This results of calculated hardening were, however, lower than measured hardening (1.46, 1.12 and 0.27 GPa for 250, 380 and 300/450 °C, respectively). In particularly, it can be noted that the difference in these three irradiation conditions is not only depend on displacement dose but also depend on irradiation temperature.

**Summary**

Reduced activity ferritic/martensitic (RAFM) steels are evolutions. Especially, high energy neutron irradiation effect is key point to develop these steels. The hardening under irradiation at low temperature may cause undesirable materials degradation. On the other hand, there is no fusion neutron can be used, in the case, high energy heavy ion was suggested to simulate the irradiation induced damage cascade. And the irradiation induced mechanical properties and microstructures were
In this work, we were mainly focused on Fe-8Cr model alloy, F82H-ODS steel. Some other materials were also used in comparison. The results of the present study are summarized as follows.

For Fe-8Cr model alloy, irradiation were conducted at 100, 250 and 300 °C in single and dual beam modes with 10.5 MeV Fe$^{3+}$ and 1.05 MeV He$^+$. It can be concluded that firstly, irradiation caused hardening at each temperature with the increase of irradiation dose. The log-log plots of irradiation hardening as a function of irradiation dose showed that there is a tendency towards saturation in hardening with the decreased temperature. Moreover, microstructure showed that small dislocation loops were the main irradiation damage in both single and dual beam irradiation up to 10 dpa. Moreover, mechanical and microstructure tests on unirradiated steel indicated that nano-hardness versus the root of dislocation density plot had a linear relationship.

F82H-ODS steel was irradiated at two constant temperatures, 250 and 380 and varied temperature 300/450 °C to 20 dpa with single ion beam. Firstly, irradiation hardening was detected, and the lowest hardening was confirmed at the variable temperature. Microstructure indicated that defect clusters were the main damage structure. Defect clusters formed close to oxide particles and intrinsic lath boundaries and dislocation lines have also been confirmed in this ODS steel. It is hinted that these initial microstructures can act as trapping and recombination sites for point defects.

In order to get the relationship between the microstructure and nanohardness, the traditional disperse obstacle model was applied. The irradiation induced microstructure, the irradiation induced nanohardness change and the dispersed barrier hardening model were considered and discussed, respectively. It indicated that invisible defect clusters should response for the difference between the calculated and measured hardening.

**Publication**
