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Characterization of Microstructure and Stability of Precipitation in SIMP Steel Irradiated with Energetic Fe Ions *

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A type of home-made reduced activation martensitic steel, high silicon (SIMP) steel, is homogeneously irradiated with energetic Fe ions to the doses of 0.1, 0.25 and 1 displacement per atom (dpa), respectively, at 300°C and 1 dpa, at 400°C. Microstructural changes are investigated in detail by transmission electron microscopy with cross-section technique. Interstitial defects and defect clusters induced by Fe-ion irradiation are observed in all the specimens under different conditions. It is found that with increasing irradiation temperature, size of defect clusters increases while the density drops quickly. The results of element chemical mapping from the STEM images indicate that the Si element enrichment and Ta element depletion occur inside the precipitates in the matrix of SIMP steel irradiated to a dose of 1 dpa at 300°C. Correlations between the microstructure and irradiation conditions are briefly discussed.

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Martensitic steels are promising candidate structural materials for the accelerator driven system (ADS) facilities with lead bismuth eutectic (LBE) as coolant due to their low activation, excellent mechanical properties and maturity as an industrial material.[1−4] In the last few years, a number of works have been reported and reviewed on reduced activation ferritic/martensitic steels such as JLF, F82H, EUROFER97 and CLAM steels.[5−8] As one candidate structural material for ADS project in China, a new type of reduced activation martensitic steel, the novel high silicon steel (named SIMP steel), has been cooperatively researched and developed by both Institute of Modern Physics and Institute of Metal Research, Chinese Academy of Sciences (CAS). In recent years, a number of studies were carried out to investigate radiation damage effect of SIMP steel with different energetic irons. Radiation effects on SIMP steel including cavity swelling, radiation induced defects(RID) and tensile behaviors in static lead bismuth eutectic(LBE) have been examined in the previous studies.[9−12] To date, numerous investigations on SIMP steel have been carried out and many experimental data are available. However, the irradiation effects on interstitial type defects including defect clusters and alloying element precipitates in SIMP steel are only slightly understood, and especially the diffusion mechanism of silicon element is not yet clear.

In this work, the SIMP steel samples were irradiated with multi-energetic Fe ions to the doses of 0.1, 0.25, 1 displacement per atom (dpa), respectively, at 300℃ and 1 dpa, at 400℃. The objective of the present work is to provide the useful information on understanding of interstitial type RID and alloying element precipitation in SIMP steel.

The material employed in the present experiment was the newly developed SIMP steel obtained from Institute of Metal Research, CAS, with a size of 15 mm × 15 mm × 1 mm. The major constituents (wt%, bal. Fe) of the SIMP steel are Cr 10.24, C 0.22, Mn 0.52, Ta 0.12, W 1.44, V 0.18 and Si 1.22. The thermal treatment procedure of the SIMP steel was normalized at 1050℃ for 30 min, and then tempered at 760℃ for 90 min. One surface of each sample was mechanically ground with sandpapers of varying grits and finally to a fine mirror finish. In the end, all the samples were ultrasonically cleaned in acetone and ethanol step-by-step.

The Fe-ion irradiation was performed in a high-temperature irradiation creep terminal chamber of the sector focusing cyclotron (SFC) in the National Laboratory of Heavy Ion Research Facility of Lanzhou (HIRFL), China. The SIMP steel samples were 3D homogeneously irradiation in the depth range of 3–20 μm using 353 MeV Fe ion transmitting through a degrader wheel with 36 Al foils of variable thicknesses. An X–Y magnetic scanning system was used to obtain a well-distributed ion beam bombarding with an homogeneity better than 95%, and the beam size was $30\times30\,\mathrm{mm}^2$. An aperture made of steel alloy with size of $15\times15\,\mathrm{mm}^2$ was placed in front of the samples to ensure the final uniformed beam with size of $15\times15\,\mathrm{mm}^2$ on the SIMP samples. With the stable beam current densities of $0.18\,\mu\mathrm{Acm}^{-2}$, the Fe-ion-irradiated fluence was approximately $6.4\times10^{10}\,\mathrm{ion\cdot cm}^{-2}$ per second and the average damage cross section of $6.5\times10^{-17}\,\mathrm{dpa\cdot cm}^2/\mathrm{ion}$ was selected, as shown in Fig. 1. Thus the corresponding displacement damage rate was $4.2\times10^{-6}\,\mathrm{dpa\cdot s}^{-1}$ in this Fe-ion irradiation experiment. The current production of the displace-

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ment damage was calculated by SRIM2008[13] for a displacement threshold energy of 40 eV, as shown in Fig. 1. The temperature distribution on the sample during irradiation was monitored by two thermocouples mounted at the side and back of the sample controlled intelligently by the heating systems. The measured maximum change of the sample temperature at 673 K was less than 5℃ under the condition of the Fe-ion beam turning on and shutting down processes.

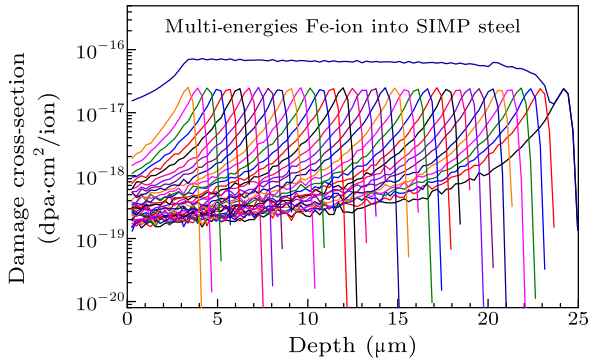

Fig. 1. SRIM calculation of the damage cross-section profile produced by 353 MeV Fe ions passing through a degrader wheel with 36 Al foils of variable thicknesses in SIMP steel.

The cross-sectional TEM samples were prepared by mechanical polishing down to approximately 30 μm in thickness. Then ion milling by using a Gatan precision ion polishing system (Gatan 691, with two Ar ion beam of 5 keV gradually decreasing to 4 keV, incident at the glancing angles of 5° for 1 h and then decreasing to 3° to minimize radiation damage and beam heating effect induced by the Ar ion) was employed to obtain the TEM foil with thickness that is transparent for electrons. The defects induced by Fe-ion irradiation in SIMP specimens were examined using an FEI Tecnai F20 TEM equipped with a double-tilt goniometer stage and operated at 200 kV with a field emission gun. The most often used image conditions were bright field (BF) and weak beam dark-field (WBDF) at $g(5g)$, $g = 110$ near $z = 111$.[14] The size and number density of defect clusters were quantifying using only the TEM micrographs of $g(5g)$, $g = 110$ near $z = 111$ for all the specimens. The thickness data of the TEM specimens in the interested region was obtained with a fluctuation of about ±10% by the contamination-spot separation method, as described in Ref. [15]. However, it should be noted that the real density of interstitial defect clusters should be larger than those obtained from the images taken under a single image condition. Furthermore, scanning transmission electron microscopy (STEM) and EDX measurements were performed to obtain detailed evidence for alloying element precipitation in the sample irradiated under different conditions.

The typical microstructure of the un-irradiated SIMP steel is shown in Fig. 2. It is observed obviously that the SIMP steel has a typical tempered martensitic lath structure containing dislocations in lath matrix and different carbide precipitates formed at a variety of boundaries and matrix. The $M_{23}C_6$-type carbide precipitates are identified mainly along martensite lath boundaries and prior austenite grain boundaries. The sizes of precipitates vary from few tens of nm to hundreds of nm. The white dashed-dotted line overlaid on Fig. 2 shows a packet boundary of prior-austenite grains in the SIMP steel.

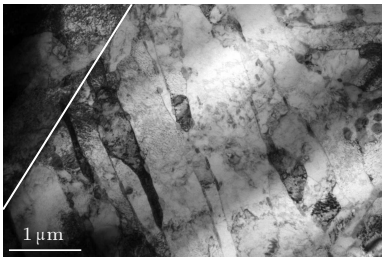

Fig. 2. Bright field images show the typical microstructure of the as-received SIMP steel. The white dashed-dotted line overlaid on the image shows a packet boundary of prior-austenite grains in the SIMP steel.

Figure 3 presents a comparison of the defect clusters or loops induced by irradiation in the SIMP steel irradiated to 0.1, 0.25, 1 dpa, at 300℃ and 1 dpa, at 400℃, respectively. The typical bright field microphotographs of defect clusters or loop formation in the SIMP steel are illustrated in the upper row of Fig. 3. The corresponding weak beam dark field images in inverted-contrast images are shown in the lower row of Fig. 3.

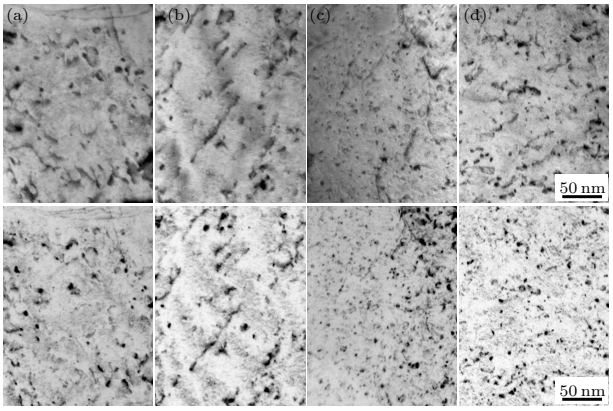

Fig. 3. Bright field images (upper row) and the corresponding weak beam dark field images in inverted-contrast images (lower row), showing the defect clusters or loops in the SIMP steel irradiated under different conditions: (a) 0.1 dpa, 300℃; (b) 0.25 dpa, 300℃; (c) 1 dpa, 300℃; and (d) 1 dpa, 400℃.

The main feature of defects induced by irradiation is small defect cluster, the black dots or small loops, which can be seen in Fig. 3. The microstructure of the SIMP steel irradiated to low doses of 0.1 and 0.25 dpa, at 300℃ including only some distortion dislocation lines and some sparse black dots are observed, as shown in Figs. 3(a) and 3(b). The density of the black dot is quite low at low dose. With increasing the irradiation dose to 1 dpa, dense small defect clus-

ters or loops are observed in Figs. 3(c) and 3(d). The results indicate that the dose threshold value of defect cluster formation in the SIMP steel is in the range of 0.25–1 dpa for the present Fe-ion irradiation. Figure 4(a) shows the temperature dependence of the density and size of defect clusters or loops in the SIMP steel irradiated to the dose of 1 dpa at 300℃ and 400℃. The corresponding size distribution of defect clusters or loops induced by irradiation are also plotted in Fig. 4(b). At the dose of 1 dpa and the irradiation temperature of 300℃, the defect cluster mean size is 3.1 nm with the density of $4.5 \times 10^{22}\,\mathrm{m}^{-3}$. When the irradiation temperature increases to 400℃, the mean size of the cluster obtained from TEM image is 4.5 nm with the density of $2.2 \times 10^{22}\,\mathrm{m}^{-3}$. This demonstrates that the size of defect clusters increases with the irradiation temperature while the defect cluster density drops quickly. Although at the same dose of 1 dpa, it is clear that both the size and density are quite sensitive to the irradiation temperature.

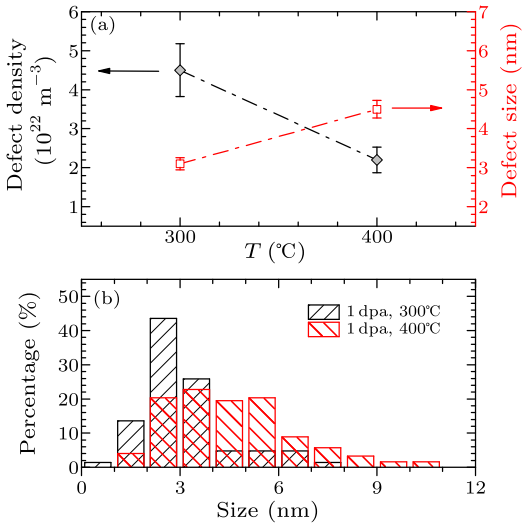

Fig. 4. (a) The temperature dependence of the density and size of defect clusters in the SIMP steel irradiated to the dose of 1 dpa. (b) The size distribution of defect cluster or loops in the SIMP steel irradiated to the dose of 1 dpa at different temperatures.

In most cases, the self-interstitial atoms caused by irradiation can migrate and accumulate much more easily at higher temperature because of the lower activation energy and migration energy and result in formation of the larger size defect cluster or loop with lower density. The present results are more or less consistent with the temperature dependence of defect cluster size and density in the previous study on T91 steel irradiated in the Swiss spallation neutron source Target-3.[14] Generally, both vacancies and interstitials are produced in equal numbers at the beginning of the irradiation, and most of them will be lost either by recombining each other or by absorption into dislocations or boundaries due to the sink effect. On the one hand, the vacancy type defects are stabilized by residual gases in the material and lead to formation of the cavity embryos. On the other hand, the dislocations, acting as biased sinks for preferential absorption of interstitials, can absorb interstitials continuously and then result in the formation of loops. Whilst the enhanced inputting of interstitials into the dislocations or loops occurred, this will cause the interstitial loops expansion, coalescence and final incorporation into the dislocation network.[16] Figure 5 presents the low magnification images of the dislocation networks in the SIMP steel after the doses of 0.25, 1 dpa, at 300℃ and 1 dpa, 400℃, respectively.

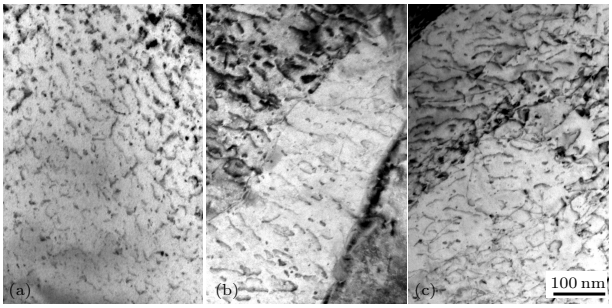

Fig. 5. Low magnification images show the dislocation networks in the SIMP steel irradiated under different conditions: (a) 0.25 dpa, 300℃; (b) 1 dpa, 300℃; and (c) 1 dpa, 400℃.

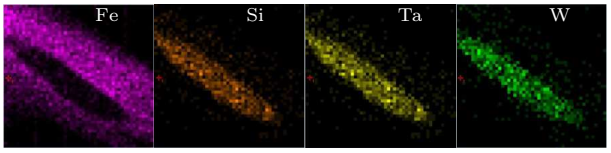

Fig. 6. The corresponding elemental chemical mapping image of a precipitate particle in the received SIMP steel.

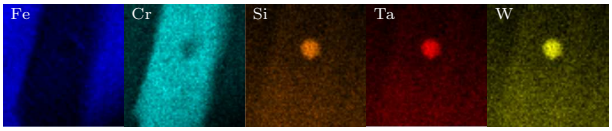

Fig. 7. The corresponding elemental chemical mapping images of an MX precipitate particle formation inside an $M_{23}C_6$ precipitate in matrix irradiated to 0.25 dpa at 300℃.

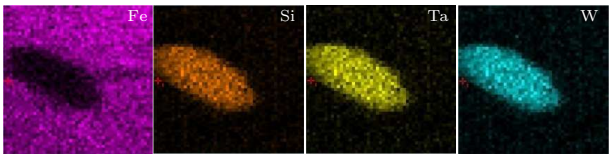

Fig. 8. The corresponding elemental chemical mapping images of an MX precipitate particle in the SIMP steel irradiated to 1 dpa at 300℃.

The sink effect of the second-phase particle, including both $M_{23}C_6$ and MC precipitates, on the cavity swelling in RAFM steel has been investigated already in our previous publication.[17] In the present work, the radiation-induced segregation (RIS) effect in the SIMP steel, as a high silicon content (up to 1.22 wt%) steel, was also analyzed using STEM and EDX measurements to obtain detailed information for alloying element precipitation. Figure 8 illustrates the STEM

image on elemental chemical mapping of a precipitate particle in the as-received SIMP steel. As we all know, the second-phase particles are introduced intentionally by prior thermal treatment to strengthen the matrix and grain boundaries. Commonly, the precipitates are all carbide and the precipitates are rich in Cr, Ta and V elements in the RAFM steel. However, because of the high silicon content in the SIMP steel, the element C will be replaced by Si usually and will result in the formation of the silicide, as shown in Fig. 6. The precipitates are rich in Si, Ta and W with low Fe content in the received SIMP steel. Figures 7 and 8 show the corresponding elemental chemical mapping images of precipitate particle in the SIMP steel irradiated to 0.25, and 1 dpa, at 300℃, respectively. Compared with the precipitates in the received SIMP steel, there is no significant change in compositions of the precipitates in irradiated SIMP steels. However, the content of the alloying elements such as Si and Ta changes with increasing the irradiation dose. EDX analysis shows that the content of the element Si increases from 2.6 wt% under the un-irradiated condition to 5.4 wt% in irradiation to 1 dpa at 300℃, and the element Ta decreases from 26.3 wt% to 16.9 wt%. The obtained values of Si and Ta content in precipitate are the average values of EDX elements line scanning measurements in the same area many times. In irradiation environments, silicon element, with a lower activation energy for diffusion in steels, can migrate easily in steel by vacancy or interstitial exchange mechanisms. With increasing the dose, it is assumed that the exchange behavior between Si with Ta will occur around the Ta-rich precipitate particle. Garner and Wolfer[18] reported that the element Si not only has a large diffusivity but also shows an increase in the diffusivity of the solvent atoms as well. The similar phenomena of the element Si enrichment and element Ta depletion inside the irradiated precipitates were observed in the present work. However, it appears that the data presented are very limited to make a definite conclusion. More detailed study is on the way.

In summary, we have investigated microstructures of the SIMP steel, as a high silicon content steel, homogeneously irradiated with energetic Fe ions by TEM. Interstitial defects and defect clusters induced by Fe-ion irradiation are observed in all the specimens under different conditions. The behaviors of alloying elements are also analyzed using STEM and EDX measurements and result in the Si element enrichment, and Ta element depletion occurs inside the irradiated precipitates.

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