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EFFECTS OF PULSED AND/OR DUAL ION IRRADIATION ON MICROSTRUCTURAL EVOLUTION IN A Ti AND Si MODIFIED AUSTENITIC ALLOY*

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The influence of pulsed 4 MeV Ni-ion bombardment, with and without simultaneous helium injection, at 958 K and damage levels from 1 to 50 dpa has been studied in a low swelling, Ti- and Si-modified austenitic stainless steel. Compared to continuous irradiation, pulsing caused an increase in the number density of interstitial loops formed during irradiation. Helium also increased the nucleation of interstitial loops. The main precipitates formed were a large number of small TiC particles uniformly distributed in the matrix, and a small number of relatively large eta and G precipitates. These coarse precipitates were somewhat larger in the pulsed specimens. Pulsing appeared to produce no significant change in swelling compared to continuous irradiation. However, for one specimen irradiated to 54 dpa, pulsing concurrent with substantial temperature fluctuations caused by beam heating may have been responsible for a larger swelling compared to continuous irradiation.

1. INTRODUCTION

Pulsed reactor operation has been considered in some magnetic fusion reactor designs. Pulsing entails intervals of irradiation where many point defects are produced, followed by off periods in which point defects anneal out thermally. The pulsing, then, could possibly modify the kinetics of the damage process and lead to a different evolution of radiation-induced microstructures and to differences in properties of first wall or structural materials.

Experimental data^{1,2} shows that changes in microstructures of continuously irradiated materials are strongly affected by helium, which is produced to levels up to thousands of parts per million in fusion reactor materials through (n,α)-reactions between high energy neutrons and component elements. Helium induces differences in nucleation kinetics of dislocation loops and cavities and also in growth kinetics indirectly through change of the sink strengths and possibly the system bias

for point defects. The strong interaction between helium and point defects as well as the changes in sink strengths may also affect the precipitation and solute segregation processes under irradiation.^{1,3}

Pulsed irradiation experiments on stainless steels have been reported previously.⁴⁻⁶ However, these used simple alloys where phase instability was not exhibited during irradiation. In another experiment, the influence of pulsing, with and without helium, on evolution of microstructures was investigated in a Ti- and Si-modified stainless steel where phase instability and solute segregation are important factors for determining the radiation-induced changes in properties.⁷ In the present experiments, a somewhat different Si and Ti-modified alloy was investigated to further assess the effects of pulsing on damage microstructure.

2. EXPERIMENTAL PROCEDURES

The composition (wt %) of Ti- and Si-modified austenitic stainless steel (F7) used

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in this study was Fe-17Cr-14Ni-1.9Mo-2.0Mn-1.1Si-0.14Ti-0.06C. Discs 3-mm diameter $\times$ 0.35-mm thick were solution-annealed for 15 min at 1373 K in a helium atmosphere. Specimens were bombarded with 4 MeV Ni⁺⁺ ions alone or simultaneously with 0.2–0.4 MeV He⁺ ions at ~958 K to damage levels of about 1 and 50 dpa using the ORNL dual ion beam Van de Graaff facility. The irradiation temperature corresponds to the maximum swelling temperature of this alloy when irradiated with steady dual beams.⁸ The relatively low dose rate of 2×10^{-3} dpa/s was employed in order to limit temperature variations caused by beam heating to less than 4 K (the usual value is 6×10^{-3} dpa/s). The pulsing schedule was 60 s on/60 s off, and the helium injection rate was 20 appm He/dpa. A few specimens, however, showed temperature oscillations of about 50 K, probably due to loose clamping against the heat sink. Details of the bombardment facility and procedures have been given elsewhere.⁹

Following ion bombardment, the specimens were electrochemically back-thinned to the peak damage depth of ~0.7 μ m. JEM 120CX and JEM200A transmission electron microscopes were used for TEM examination.

3. EXPERIMENTAL RESULTS

3.1. Dislocation loops

Dislocation loops produced in F7 irradiated continuously with Ni ions and dual beams of Ni and He ions are shown in Figure 1(a) and (b), respectively. The loops formed during pulsed irradiation with Ni ions are shown in Figure 1(c). These loops have stacking fault fringes and were identified as interstitial type. There are significant differences in the sizes of the loops, as shown in Figure 2. Pulsed irradiation reduced the overall size of the loops and, in addition, caused a notable increase in the number of very small loops with

diameters less than 50 nm. The loop densities were 4×10^{19} and $2 \times 10^{19}/\text{m}^3$ for pulsed and continuous nickel ion irradiation, respectively. Continuous dual beam irradiation increased the loop number density to $7 \times 10^{19}/\text{m}^3$. These dislocation loops grew and interacted to form a dislocation network with increasing dose. The saturation dislocation density, 1 to $2 \times 10^{14} \text{ m}^{-2}$ did not depend strongly on the irradiation mode.

3.2. Cavity formation

Cavities were formed only in specimens irradiated with both Ni and He ions, regardless of whether the irradiations were continuous or pulsed. For pulsed irradiation at ~40 dpa, cavities were small and swelling negligible both for one specimen that underwent temperature excursions between 947 and 907 K caused by beam heating as well as for another specimen which remained at 942–945 K. However, above 50 dpa swelling increased and three types of cavities were observed. Large (70 to 100 nm diam) voids were attached to large precipitates while an intermediate class (10 to 50 nm) and a class of small cavities (<10 nm) were found in the precipitate-free matrix. The small cavities are thought to be gas bubbles and the larger ones to be voids.¹⁰ As seen in Figures 3(a) and 3(b) the ~50 dpa specimen which experienced both flux and 60 K temperature oscillations contained more and larger voids than a continuously-irradiated specimen. The specimen which had the flux and temperature oscillations exhibited a swelling of 1%, several times that of the steady-irradiated specimen.

3.3. Precipitate formation

Figure 4 shows the cavity and precipitate features of F7 for continuous and pulsed irradiation with or without helium. Very small MC particles (e.g., Figures 4a and 4c), which were identified from dark field electron micrographs,

FIGURE 1

Dislocation loops produced in F7, (a) continuous, Ni-ions (0.5 dpa, 951 K), (b) continuous, dual beam (0.8 dpa, 955 K), and (c) pulsed, Ni ions (0.7 dpa, 956/954 K). Very small loops are seen as spots in (c).

FIGURE 2

Size distribution of dislocation loops in F7 irradiated (a) continuously with Ni ions (0.5 dpa, 951 K) and dual beams (0.8 dpa, 953 K) and (b) pulsed with Ni ions (0.7 dpa, 956/954 K).

FIGURE 3

Cavity microstructures of F7 irradiated with dual beams (a) continuously (52 dpa, 968 K) and (b) pulsed (54 dpa, 958/900 K).

FIGURE 4

Dislocations and precipitates in F7 irradiated continuously with (a) Ni ions (34 dpa, 943 K) and (b) dual beams (52 dpa, 968 K), and by pulsing with (c) Ni ions (39 dpa, 945/943 K) and (d) dual beams (40 dpa, 945/942 K).

were found in specimens irradiated to a total dose of about 50 dpa for all irradiation conditions. There were also larger particles, eta and G phases (as identified by diffraction patterns) present in the specimens irradiated with dual beams for both continuous and pulsed modes, Figures 4(b) and 4(d). These micrographs also show that pulsing apparently formed eta particles of greater size and lesser number density than those formed during continuous irradiation.

4. DISCUSSION

The nucleation of interstitial loops occurs in the early stages of irradiation. Pulsed irradiation repeats this process at the start of every cycle. Therefore, new loops nucleate successively during pulsing. Thus, pulsing resulted in a large loop population including the very small loops which were nucleated in more recent pulses. During the beam-off period, vacancies which were present at steady state during the on period can migrate directly to sinks without recombination with

interstitials, because the interstitial concentration drops rapidly when the beam is turned off. Therefore, some portion of the vacancies may flow to the interstitial loops and erode them. The same observations of smaller interstitial loops resulting from pulsing was reported previously,⁷ with the same explanation being proposed. In both cases the total loop area was reduced (sevenfold in this study, two to eightfold in the preceding study) for pulsed relative to steady irradiations, which is consistent with the above explanation. The interstitial loops were also notably refined by dual beam as compared with single ion irradiation. This may be due to helium tying up vacancies which would otherwise eventually erode the high number of loops nucleated at the beginning of an irradiation.

Precipitation in this alloy began with fine MC particles which give way to large eta (and some G) precipitates by about 40 dpa. Pulsing yielded larger eta precipitates, presumably because eta phase can develop upon annealing,¹¹ which occurs here in the beam-off periods. The

progression from MC to (mainly) eta phase resembles the observations of the earlier investigation in a similar alloy⁷ except that there the MC regime persisted to higher doses (above 70 dpa) for pulsed irradiation. Such a difference in phase evolution is presumably due to the fact that the MC particles are principally TiC and there is less titanium (0.14 wt %) in the current material compared to the former (0.2 wt %). The TiC particles thus should not dominate the microstructure as long in the present alloy and formation of the massive phases would not be so retarded.

The appearance of swelling seems to be linked to the formation and growth rate of the eta and G phase precipitates, because these precipitates have been shown to collect point defects effectively and thus enhance the growth rate of voids attached to them.^{12,13} Thus, the specimens exhibiting the highest swelling contained large cavities associated with large precipitates as seen in Figure 3(b). During the initial part of each beam-off period, the same unopposed vacancy flow that erodes interstitial loops (above) ought to augment cavity sizes. For the special case of pulsing accompanied by beam-off temperature drops, an additional cavity growth factor could ensue from acceptance of surplus thermal vacancies engendered by the temperature drop. These vacancy condensation mechanisms should only have a lasting effect, however, for cavities that are above the critical size for bias-driven growth and are therefore free to retain their absorbed vacancies during the next beam-on period. Otherwise, the cavities would have to again decrease to their stable size during the next beam-on period or indeed the postcondensation remainder of the beam-off interval.¹⁴ This qualification may explain why similar

temperature variations failed to cause the same changes at 40 dpa, where cavities were still small and probably subcritical, that they did at 50 dpa.

5. SUMMARY

A study of the phase stability of a Ti-and Si-modified stainless steel under pulsing with and without helium has been performed. The pulsing causes an increase in loop number density. The presence of helium also promotes loop nucleation. Helium is essential for cavity formation. Pulsing did not greatly alter cavity formation. However, one specimen which experienced ~50°C downward temperature fluctuations during beam-off periods showed substantially higher swelling than specimens subjected to continuous irradiation. Precipitates of MC, eta, and G-phase were formed during these irradiations, with most of the MC disappearing at the higher doses. Pulsing enhanced somewhat the formation of eta phase particles with respect to continuous irradiation. These results have been interpreted according to present understanding of radiation-enhanced precipitation and precipitation-enhanced cavity swelling.

The effects of pulsing on loop formation are similar to those observed earlier in a similar alloy.⁷ The lesser effects of pulsing on phase stability and swelling in the present case are attributed to the significantly lower concentration of Ti. This reduces the enhancement of TiC by pulsing that was observed in the earlier study and therefore diminishes the associated delay in the formation of the radiation-enhanced coarser G and eta phases, and in the consequent swelling by precipitate point defect collection.

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