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THE INFLUENCE OF HELIUM ON MICROSTRUCTURAL EVOLUTION IN AN ION-IRRADIATED LOW-SWELLING STAINLESS STEEL*

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The evolution of damage in a low-swelling stainless steel under ion irradiation is examined. The influence of the presence of helium and its mode of injection upon dislocation evolution, phase instability, and swelling are investigated. Comparison of the response of the low-swelling alloy with that of a high-swelling alloy leads to some conclusions on the origin of the observed swelling resistance. The dislocation loop evolution can be modified by the presence of helium during the nucleation stage. The influence of helium on phase instability arises from its modification of the loop substructure at which solute segregation occurs. Phase instability is not a sufficient condition for void formation in the swelling resistant alloy. Titanium getters soluble gases which aid void nucleation, while silicon may influence swelling by a trapping mechanism.

1. INTRODUCTION

It has long been recognized that helium and/or other gases play a critical role in void nucleation [1,2]. For this reason, helium has been injected in simulation irradiations in an attempt to obtain a closer match to neutron irradiation [3–5]. In addition, ion irradiation studies, in which many irradiation parameters can be controlled independently, have been applied to understanding the mechanisms of swelling resistance and other irradiation produced processes, such as solute segregation or phase instability. While helium is injected to aid void nucleation, its presence may also affect the other components of the damage structure; dislocation loops and segments, and precipitate phases. In addition, the mode of helium implantation can produce marked changes in the influence that helium has upon the damage structure [5–8].

The present investigation deals with the influence of helium on the evolution of the damage structure in an ion-irradiated, modified 316 stainless steel alloy, LS1A. This alloy is highly swelling resistant under ion irradiation in the absence of helium, apparently as a result of a high barrier to void nucleation [9]. The alloy also exhibits solute segregation to dislocation loops and phase instability of the austenite matrix under ion irradiation [10]. The influence of simultaneously injected helium on the damage evolution has been examined for two He/dpa rates, and the influence of cold preinjection of helium has been investigated. Preliminary results will be presented. A more comprehensive paper will be published later.

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2. EXPERIMENTAL PROCEDURE

The alloys were arc melted from high-purity constituents and the resulting chemistries are given in Table 1. Specimens were 3-mm-dia disks punched from 0.75 mm sheet and final annealed at 1050°C for 1 h under argon. Disks were vibratorily polished with various abrasives down to 0.1 μm diamond abrasive. Irradiations were performed using 4 MeV Ni ions on the ORNL dual-beam Van de Graaff facility. Some specimens were simultaneously injected with helium at either 0.2 or 20 appm He/dpa. Other specimens were preinjected at 25°C to various helium levels and subsequently nickel ion irradiated. Bombardments were performed at 625°C to exposures between 1 and 70 dpa at a dose rate of $\sim 7 \times 10^{-3}$ dpa/s. Step-height measurements were made relative to an unirradiated area of the specimen. Disks were sectioned and back-thinned to the peak damage depth ($\sim$0.7 μm). Conventional transmission electron microscopy (CTEM) and analytical electron microscopy (AEM) were performed on a JEOL 100 C with Kevex energy-dispersive spectroscopy (EDS) capability.

3. RESULTS

3.1 Irradiations without helium injection

To provide a basis for comparison, the damage evolution in uninjected LS1A and a nominal 316 stainless steel, G7, will be summarized from a more detailed paper [11]. At low dose ($\sim$1 dpa), the two alloys exhibit similar structures: predominantly faulted interstitial loops, with some isolated perfect loops and dislocation segments. However, there are quantitative differences; the average loop diameter in LS1A is roughly twice that of G7, while the loop densities in G7 are two to threefold higher. At 1 dpa, faulted loops in LS1A are $\sim$430 Å in diameter at a density of 5×10^{14} cm^{-3}. At this dose, solute segregation of silicon and nickel is observed in LS1A as previously reported [9]. Any segregation in G7

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Table 1. Alloy Compositions (weight percent)

Alloy	Cr	Ni	Mo	Si	Mn	Ti	Al	Nb	V	Co	Zr	W	Cu	C
G7	16.5	13.7	1.86	0.44	2.05	<0.01	<0.02	<0.005	<0.005	<0.005	<0.005	0.03	0.01	0.04
LS1A	16.4	13.7	1.73	1.05	2.05	0.15	<0.02	0.04	0.04	<0.005	0.06	0.03	0.05	0.08

is below the detection limit of the technique applied. As the damage level increased, the dissimilarity of the damage structures increased. In G7, loops nucleate, grow, unfault, and become part of the dislocation network, which increases in density with dose. In LS1A, loop unfaulting apparently occurs at a very slow rate. Between 3 and 10 dpa, irradiation-induced precipitates appear to replace loops, the loop density drops, and a corresponding increase in the network density occurs. Two precipitate types occur; both are cubic phases with lattice parameters ~11 Å. One precipitate is isostructural to $M_{23}C_6$, the other precipitate to M_6C. Both phases are rich in silicon and nickel relative to the matrix. No phase instability is observed in G7 at any dose. Below 30 dpa, voids are nucleated in G7 and swelling reaches 4.2% by 70 dpa. In LS1A (Fig. 1a), the degree of phase instability increases with dose until saturating at ~5% volume fraction at 70 dpa. At this point the matrix silicon concentration has been reduced to <60% of the original level. No void nucleation is observed in uninjected LS1A to dose levels of 600 dpa, at which the swelling in G7 is estimated from step-height measurements as ~110%.

3.2 Simultaneous helium irradiations

With the simultaneous injection of helium into these alloys, significant changes in the damage structures are observed at higher doses (10–70 dpa), while little change occurs at low doses (~1 dpa) to the interstitial clustering in dislocation loops. At both 0.2 and 20 appm He/dpa, the average loop diameter in LS1A at 1 dpa is ~400 Å and the loop density is 2–3 × 10^{14} cm^{-3}. The loop density for the 20 appm He/dpa irradiation is only slightly greater. Under simultaneous 0.2 appm He/dpa irradiation, both alloys follow the same early stages of evolution which they exhibited in the uninjected condition. However, voids are observed in both alloys at 10 dpa. In G7, the voids are ~250 Å diameter and uniformly distributed at 1.7 × 10^{14} cm^{-3}. The 350 Å voids in LS1A are inhomogensously distributed at ~6 × 10^{12} cm^{-3} in association with irradiation induced precipitates. The inverse is not true in general. By 70 dpa the voids in G7 have grown to ~770 Å diameter and their density has doubled, resulting in a void swelling of 8.7%, twice that for uninjected G7. In contrast, the voids in LS1A have continued to nucleate to a density of 1.5 × 10^{14} cm^{-3} and grown to ~710 Å for a swelling of 3.5% (Fig. 1b). At the higher implantation rate of 20 appm He/dpa, the cavity nucleation rate in LS1A is increased. At 10 dpa, the cavities are 200 Å in diameter, uniformly distributed at 6 × 10^{13} cm^{-3}, and are still in association with the irradiation-produced precipitates. At 70 dpa (Fig. 1c), the increased nucleation produces ~2.5 times higher cavity number density (3.6 × 10^{14} cm^{-3}), a significant decrease in size (430 Å), and a factor of ~2 decrease in swelling (1.8%) relative to the 0.2 appm He/dpa case. (Calculations show that sufficient injected helium is present in this case for the cavities to be equilibrium bubbles.) The assessment of the degree of phase instability in LS1A under the three preceding irradiation conditions is difficult as a result of the association with voids. It appears that the precipitate size decreases, while their number density increases slightly when going from uninjected to 0.2 appm He/dpa irradiation and finally to 20 appm He/dpa. The general impression from Fig. 1 is that the degree of phase instability decreased with increasing amounts of simultaneously injected helium.

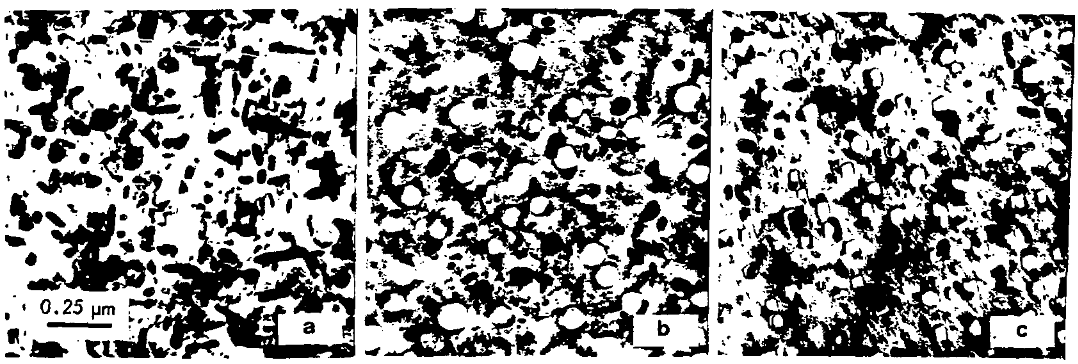

Fig. 1. Damage structure of LS1A irradiated to 70 dpa at 625°C. (a) Uninjected; (b,c) simultaneous injection; (b) 0.2 appm He/dpa; (c) 20 appm He/dpa.

3.3 Preinjected helium irradiations

Room-temperature preinjection of helium at levels from 14 to 112 appm results in significant modification of the damage evolution at both both low- and high-dose levels. In both 14 appm preinjected alloys at 1 dpa, the loop structures have been refined in scale; the loops are smaller in size, but are present at higher number densities. The average loop diameter in LS1A is 90 Å at an apparent density of $\sim 2 \times 10^{15}$ cm^{-3}. Actual loop densities are probably higher as the result of unresolvable defects. At 10 dpa, the damage structures of both G7 and LS1A are still composed primarily of loops. No voids are observed in either G7 or LS1A and little or no phase instability is observed in LS1A. Upon irradiation to 70 dpa, the changes in damage evolution remain apparent. Both alloys still exhibit significant loop populations, which is not observed in either the uninjected or simultaneously injected irradiations. In G7, preinjected helium doubled the void density (3.2×10^{14} cm^{-3}), but the void diameter decreased to 570 Å, leaving the swelling approximately the same (3.8%). LS1A exhibited only occasional 100 Å cavities; the resultant swelling could only be estimated as being less than 0.1×10^{-2}. In addition, there were indications of the formation of grain boundary cavities. No phase instability was exhibited by the preinjected LS1A at this dose.

LS1A cold preinjected to 28, 56, and 112 appm He and subsequently irradiated to 70 dpa confirmed the trends exhibited by the 14 appm He preinjected specimens. Significant populations of faulted loops still exist, and a matrix population of small cavities in the range 100–200 Å was observed (Fig. 2a). As the level of preinjected helium increased, the density of these cavities increased, possibly sixfold from the 28 appm to the 112 appm He conditions. For the higher helium levels, cavities ∿170 Å in diameter appear in bands parallel to some grain boundaries and the grain boundaries themselves contain high densities of small cavities (∿30 Å). As in the 14 appm He case, no phase instability is exhibited for these helium levels.

One other experiment was performed to aid in the understanding of the different effects of preinjected versus simultaneously injected helium. While preinjected helium affects dislocation loop evolution, phase instability and void swelling, simultaneously injected helium has little effect on loop evolution, has second-order effects on phase instability, but has major effects on void formation. After irradiation to 10 dpa, where loop nucleation is over and significant phase instability is exhibited, 14 appm He was injected into LS1A and the irradiation was continued to 70 dpa. The damage structure at 70 dpa was similar to that observed for either simultaneous irradiation (Fig. 2b). Voids ∿600 Å in diameter are observed in association with the extensive phase instability. The 1.2×10^{14} cm^{-3} density of voids results in 1.6% swelling while ∿3.2% volume fraction precipitate is present.

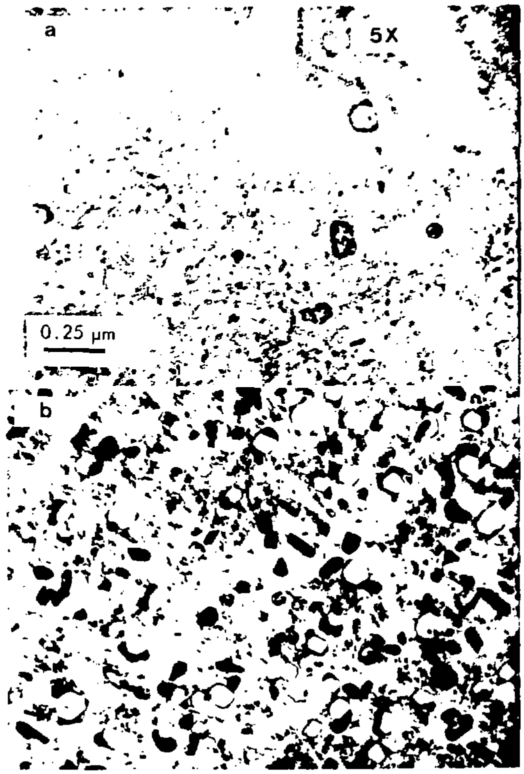

Fig. 2. Damage structures of LS1A irradiated to 70 dpa at 625°C. (a) 56 appm He, preinjected. (b) 14 appm He injected at 10 dpa.

4. DISCUSSION

A brief summary of the results will be presented to aid the subsequent discussion of the influence of helium on the evolution of the damage structure. The effects will be discussed in the order in which they appear during irradiation.

1. Loop evolution in both alloys is only slightly influenced by simultaneous helium injection. Helium preinjection significantly refines the scale of loop nucleation.
2. LS1A exhibits extensive phase instability which removes silicon and nickel from the matrix. Despite this instability, the alloy is swelling resistant in the absence of helium.
3. Phase instability in LS1A is strongly curtailed by preinjected helium, but is only slightly reduced by simultaneous injection.
4. G7 swells readily in the absence of helium; swelling increases with simultaneous helium injection, but is not changed significantly with preinjection.
5. Though 0.2 appm/dpa simultaneous helium injection results in void nucleation and swelling

in LS1A, the increased cavity nucleation under 20 appm/dpa injection decreases swelling.

6. Though helium preinjection promotes some cavity nucleation in LS1A, nucleation and/or growth are slow relative to that under simultaneous injection.

7. Helium injection after the onset of phase instability in LS1A produces a damage structure similar to that resulting from simultaneous injection.

The presence of helium apparently can modify the nucleation of interstitial dislocation loops (point 1). For the 0.2 and 20 appm He/dpa simultaneous injections, insufficient helium is present during the loop nucleation stage below 1 dpa to significantly modify their evolution. This is not the case for preinjection, where loop densities more than double. Baskes et al. [12] have calculated that there should be a strong binding of helium to both interstitials and vacancies. Johnson [13] has pointed out that the trapping resulting from such binding of both defects should result in enhanced interstitial cluster nucleation during the transient period early in an irradiation. The survival of this high density of loops into the steady-state regime depends on the behavior of the trapped vacancies. Such a mechanism may be the origin of the higher loop nucleation in the preinjected alloys. The subsequent competition between the larger number of loops for the irradiation-produced defects would decrease the loop growth rate, as is observed. The smaller loop sizes could delay loop unfaulting and interaction to form the dislocation network. Such behavior may be indicated by the survival of significant loop populations in the preinjected alloys.

A sequence of events has been proposed for phase instability (point 2) under irradiation in LS1A [10]. Silicon and nickel segregate to faulted loops under the influence of the vacancy and/or interstitial fluxes [14]. Their concentrations increase until one or both exceed some "solubility limit," and an irradiation-induced second-phase precipitates at the loop or its fault plane. Further solute segregation may occur at the precipitate, allowing further phase instability and depletion of the matrix of the segregating elements. Similar solute segregation probably occurs in G7, but is insufficient to result in phase instability. As the silicon levels of G7 and LS1A differ by a factor of ~2.5, LS1A must exhibit solute segregation under ion irradiation just slightly above that required for phase instability.

LS1A exhibits phase instability prior to or in the absence of void swelling. Under such conditions there are only two types of defect sinks — dislocations (preferential interstitial sink) and precipitates (probably neutral sink). As interstitials are preferentially absorbed at dislocations, an excess flux of vacancies would tend to arrive at the precipitates. This flux might transport helium to the precipitate interfaces. For either of these reasons, voids might be expected to nucleate in association with the precipitates. In addition, the precipitate could provide sites for heterogeneous nucleation of voids.

The swelling resistance of uninjected LS1A relative to G7 is not impaired by its extensive phase instability. Brager and Garner [15] have speculated that the depletion of silicon from 316 stainless steels by phase instability results in a loss of swelling resistance. While it may be necessary, phase instability in LS1A is not a sufficient condition for swelling (point 2). The physical association of voids and precipitates which is taken as an indication of a direct cause-and-effect relation between phase instability and swelling can be explained by the effects discussed previously.

The influence of injected helium on the phase instability of LS1A (point 3) can be understood in terms of its influence on the loop evolution. Simultaneous helium injection has little effect on loop evolution and therefore little effect on the subsequent phase instability. The slight reduction in phase instability at 20 appm He/dpa may reflect the slight increase in loop density in this case. The suppression of phase instability in the preinjected LS1A is a consequence of the higher loop density and slower loop growth. The decreased fluxes of point defects to an individual loop results in a lower degree of solute segregation. Apparently in the preinjected LS1A, the solute segregation has decreased below the critical "solubility limit" and no phase instability occurs. Thus, the matrix is not depleted of the segregating solutes.

LS1A is swelling resistant relative to G7 as a result of a high resistance to void nucleation which arises from the silicon and titanium in the alloy (point 2). Solute atoms can affect void nucleation in a number of ways, two of which are trapping and gettering. Solute atoms can trap point defects and induce higher recombination. Rate theory modeling has shown that solute trapping of either interstitials or vacancies can result in large suppressions of void nucleation and smaller reductions in void growth [16]. The silicon segregation to dislocation loops observed in LS1A is consistent with binding of silicon to vacancies and/or interstitials [14]. It is reasonable to expect silicon trapping to produce some suppression of void nucleation and growth in both alloys. Trapping may be greater in LS1A as the silicon level is 2.5 times greater.

Solute atoms can suppress void nucleation by gettering soluble interstitial gases such as nitrogen and oxygen which aid void nucleation. The swelling behavior of the uninjected alloys is consistent with such a mechanism. Sufficient soluble gases are present in G7 to nucleate voids, while in LS1A the silicon and/or titanium getter these impurities. The dependence of void nucleation and growth in LS1A on the presence of helium (points 5–7) is in agreement with the proposed gettering mechanism. While both silicon and titanium are gettering agents, titanium probably plays the dominant gettering role to result in the large differences between G7 and LS1A. The level of titanium is more than 20 times greater in LS1A, while the silicon only differs by ~2.5. In addition, the silicon level in the LS1A matrix is depleted by phase instability under irradiation.

The influence of helium on the swelling behavior of the two alloys (points 4–7) indicates the different possible effects of injection. In simultaneously injected G7, the helium results in earlier and more prolific void nucleation. At 70 dpa, the void number density is doubled and void sizes increase slightly. On the other hand, preinjection of G7 does not result in resolvable voids at 10 dpa. At 70 dpa, the void number density is the same as the simultaneously injected G7, but the swelling is only one-half. It is possible that the slow dislocation evolution in the preinjected alloy does not provide sufficient biased sinks for rapid growth until higher doses. The influence of helium on swelling in LS1A differs from that of G7 for two reasons — phase instability and gettering of soluble gases. Simultaneous helium injection provides gas to aid void nucleation. At 0.2 appm He/dpa, it appears that at least void nucleation and possibly void growth rate are lower in LS1A than in G7. This may reflect the effects of both gettering, which lowers the nucleation rate, and trapping, which reduces both nucleation rate and growth rate. The higher nucleation rate in 20 appm He/dpa injected LS1A results in a factor of 10 greater void density at 10 dpa and a factor of 2.5 greater at 70 dpa relative to the 0.2 appm He/dpa case. This results in a 50% decrease in swelling at 70 dpa. There is no significant difference in the damage structures in terms of either dislocation substructure or phase instability. One possible situation where increased void nucleation could lead to lower swelling is where the void growth kinetics change from dislocation dominant to void dominant control [17]. For such an explanation to be correct, the capture efficiency of the dislocations in LS1A must be low, since based on the measured dislocation density and void distribution and the capture efficiency of a perfect dislocations, the kinetics should be dislocation-dominated. Such a reduced capture efficiency could result from solute segregation poisoning the dislocations. A second possibility is the trapping of defects by the high concentration of injected helium. This trapping could lead to enhanced point defect annihilation and lower swelling. Lastly, the effects of the helium on the phase instability may influence the sink strength of the precipitates or the number of solute traps in the matrix. This in turn can result in reduced swelling.

5. CONCLUSIONS

1. Helium can modify loop nucleation if present in sufficient concentration early in irradiation. This effect is consistent with trapping of point defects by helium.

2. Helium can influence phase instability in LS1A by modifying the evolution of the dislocation substructure.

3. Phase instability may be a necessary condition for void swelling in LS1A, but it is not a sufficient condition.

4. In part, the swelling resistance of LS1A is the result of difficult void nucleation caused by gettering of soluble gases by titanium. Silicon may reduce swelling through a trapping mechanism and as a secondary gettering agent.

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