INFLUENCE OF DILUTE IMPURITIES ON THE EVOLUTION OF DEFECT CASCADES IN NICKEL*

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INFLUENCE OF DILUTE IMPURITIES ON THE EVOLUTION OF DEFECT CASCADES IN NICKEL

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We have employed transmission electron microscopy to study the formation of vacancy dislocation loops from defect cascades produced by a 50 keV Kr⁺ ion irradiation of Ni and its dilute alloys with Si and Al. An unusual and reproducible dependence of the loop collapse probabilities and loop size distributions on solute content was found. The results are explained by impurity caused changes in both the collisional and thermal spike (molten zone) phase of the defect cascade. Specific mechanisms include disruption of focussons by impurities to change the energy density profile within the collision cascade, interruption of replacement collision sequences, and interstitial trapping to reduce recombination during the thermal spike phase. Applying these mechanisms to a statistical distribution of the spatial extent of cascades and to the melt-zones, all the experimental observations can be explained.

1. Introduction

Developing an understanding of the basic mechanisms controlling the formation of vacancy loops from displacement cascades is of fundamental importance as they play a role in void swelling, irradiation creep and embrittlement^{1,2}. In this paper we report on the effect of adding undersized and oversized solute atoms to Ni on the vacancy loop formation probability and show how such studies allow the physics of cascade collapse to be probed.

Previous studies of solute effects on the loop formation probability in Ni show complex trends. Robinson and Jenkins³ found that the addition of ≤ 17 % Cr to Ni reduced the defect yield (which is defined as the fraction of displacement cascades that form a defect) by as much as 60% when 80 keV Ni⁺ ions were used for the irradiation. No dependence of the defect yield on the alloy content was found when 80 keV W⁺ ions were used for the irradiation. The average defect

image size increased with increasing ion mass, but no dependence on alloy concentration was found. Robinson and Jenkins³ attributed the defect yield reduction observed in the alloys irradiated with Ni^+ ions to a decrease in the loop nucleation probability in the cascade core. They suggested that the similarity in the image sizes indicated that loop growth by the accretion of vacancies was similar in all alloys.

Shoaib et al⁴ irradiated dilute ($<7\%$) Ni-Si and Ni-C alloys with 100 keV Ni^+ ions at room temperature. They found that the defect yield initially decreased as Si was added, and reached a minimum at 0.71at %Si before it increased. A similar behavior was found in the Ni-C alloys. They attributed these changes in the defect yield to the solute elements dechanneling replacement collision sequences and to them acting as heterogeneous nucleation sites for vacancy loops. As the solute content was increased the latter effect dominated and, hence, the increase in the defect yield.

In this paper we report on the effect of Si and Al additions to Ni on the loop formation probability and defect image size. These solutes were chosen as Si is undersized and Al is oversized in the Ni lattice. They are therefore expected to have different trapping strengths. The microstructure produced by a 50 keV Kr^+ ion irradiation was characterized by using transmission electron microscopy techniques. The observations are interpreted in terms of the molten-zone model for vacancy loop formation, which is based on molecular dynamic computer simulations of low-energy displacement cascades⁵⁻¹⁰. These simulations have shown that in Cu and Ni the central portion of the cascade is molten. Although loop formation was not found in the simulations, it was proposed that if the solidification rate was slow, vacancies would be formed toward the end of the solidification process and in the center of the molten zone volume⁶⁻⁸. This condition would favor loop formation. This model of vacancy loop formation has been used successfully to explain qualitatively the different defect yields in copper and nickel^{2,11,12}, the decrease in the defect yield with decreasing irradiation temperature¹² and the loss, coalescence of loops that occurs as the ion dose is increased¹².

2. Experimental Procedures.

The alloys used in this study were produced by arc-melting high-purity elements in a flowing Argon gas environment. The inductively coupled plasma analysis technique was used to determine the impurity content of the alloys, no significant level of any impurity was found. TEM discs were annealed at 900° C in vacuum for 12 hours before being jet electropolished to perforation.

Electron transparent discs were irradiated at room temperature with 50 keV Kr^+ ions. Samples of the different alloys were irradiated in the same batch, which

eliminated the need for accurate ion dosimetry. The damage structure was examined by using the transmission electron microscope. Details of the procedures used to determine the defect yield and to measure the defect image size are described elsewhere¹³.

3. Experimental Results.

In Figure 1 micrographs showing the damage structure produced in the alloys after a room temperature irradiation to an ion dose of 6×10^{11} ions cm^{-2} are compared. Qualitatively, it can be seen that the highest defect density occurs in the Ni - 0.5% Al (Figure 1(d)) and Ni- 0.6% Si (1(b)) alloys, and that the largest defects form in the Ni-7% Al (1(f)) and the Ni - 4% Si (1(c)) alloys. The dependence of the defect yield on the alloy content is quantified in Figure 2 where the defect yield is plotted as a function of the alloy content. It is worth noting that each data point was generated from at least 10 micrographs taken from two different samples and that the total error from the beam inhomogeneity and the microscopy was about 10 %. Therefore, the reported defect yield differences are outside the combined experimental errors and are significant. Figure 2 shows the complexity of alloying effects, the defect yield initially increases and then decreases as more solute is added. In the Ni-Al system, the defect yield in the high solute content alloys is less than in pure nickel.

The effect of the amount of alloying element on the defect image size is shown more clearly in the image size distributions presented in Figures 3 and 4 for the Ni-Si and the Ni-Al alloys, respectively. The distribution from pure Ni is included in both figures for purposes of comparison. From these distributions it can be seen that the average defect size increases only in the Ni-4% Si and the Ni-7% Al alloys. The average defect size was 2.15 nm in Ni compared to 2.8 and 2.59 nm in the Ni-4% Si and the Ni-7% Al alloys, respectively. Also, larger defects were present in the alloys compared to pure Ni. The largest defect observed in Ni had an image diameter between 4.5 and 5.0 nm whereas in the Ni - 4%Si and Ni-7%Al the largest defects had an image diameter between 7.0 and 7.5 nm.

The loop Burgers vector distributions were similar in all alloys. Typically, about 2/3 of the visible defect loops could be unambiguously assigned a Burgers vector. Of that population about 80 % were either Frank loops or Frank loops that had partially dissociated toward a stacking-fault tetrahedra and the remaining 20% were perfect loops.

4. Discussion

Molecular dynamic computer simulations of cascade formation have indicated that a portion of the cascade exists in a molten state for a short time⁵⁻¹⁰.

It is envisioned that as the molten zone solidifies vacancies will be created since there are more lattice sites than atoms; the atoms having been removed from the melt-zone during the displacement phase of the cascade. The distribution of vacancies within the melt-zone will depend on the duration of the molten zone, which will depend on the melting temperature of the material and the rate of solidification. For slow solidification rates the vacancies will be created toward the cascade center, which will favor loop formation.

One prediction of the molten zone model of a displacement cascade is that the duration of the molten zone is dependent on the melting temperature of the material^{2,6,8}. When the cascade dynamics are similar, the vacancy loop formation probability is predicted to be higher in the lower melting temperature material. In Cu-Ni^{14,15} and Ag-Pd¹⁵ alloys, it has been shown that the loop formation probability decreases for large increases in melting temperature. However, the relationship between the defect yield and melting temperature was not simple. For example in the Cu-Ni system, at < 20 % solute addition the change in the defect yield did not follow the expected trend. For both alloy systems used in this program, the liquidus is shallow and the largest melting temperature decrease is 25° C for the Ni - 4 at% Si alloy. This small reduction (2 %) in melting temperature is not expected to have a dramatic effect on the duration of the melt-zone and therefore on the loop formation probability. The following discussion therefore considers how other solute atom effects, such as disruption of focussons and replacement collision sequences and solute-interstitial trapping, can effect the size and duration of the molten zone and, consequently, the loop formation probability and the loop size.

The propagation of replacement collision sequences and focussons will be disrupted by the solute atoms because of the size and mass difference. The interruption of a replacement collision sequence will place the self-interstitials closer to the molten zone. If these interstitials are included in the molten zone then the difference in density between the molten zone and the matrix will be smaller and fewer vacancies will be created. This will cause a decrease in the loop size and, perhaps, the loop formation probability. Although the effect of disrupting replacement collision sequences could account for the observed decrease in defect yield with increasing solute content, it cannot account for the observed increase in defect image size.

The disruption of a focusson chain by a solute atom will cause the energy to be deposited closer to the molten zone. A consequence of this is that the temperature of the volume surrounding the molten zone will be increased (i.e., the effective energy density will be increased), which will increase the duration of the molten zone¹⁰. This in turn will promote vacancy agglomeration in the cascade

center, which will enhance loop formation. By this process the loop formation probability from all sizes of displacement cascades will be increased. The magnitude of this effect can be estimated as follows. For a solute concentration of 0.5 at%, one in every 200 atoms will be a solute atom. If the average length of a focussion chain is taken to be 30 atoms, then one focussion chain in seven will encounter a solute atom. If we assume that the solute atom is the 15th atom in the chain, the total number of atoms involved in the propagation of the seven focussions will be reduced from 210 to 195. This will reduce the effective cascade volume, which we define to include the focussion chain length, by 21%. It is not known how changes of this magnitude will influence the loop collapse probability, but in Ni where only 33% of the displacement cascades form visible loops small changes could have a profound effect.

Solute atoms can trap self-interstitials, which would prevent them from being pulled into the molten zone. This mechanism would increase the number of vacancies, which in turn would increase the defect yield and the size of the vacancy loop.

From the above discussion it is evident that if only one solute effect is considered, the experimental observations presented in section 3 cannot be accounted for. However, the formation of the displacement cascade is a statistical process and the cascades will exist with a range of volumes. It is therefore necessary to consider the consequences of the different solute atom effects on this distribution. In considering these effects, it is assumed that the total volume of the molten zone is independent of the volume of the displacement cascade. Justification for this assumption is provided by the work of Caro et al ¹⁶, who demonstrated that the molten zone volume was dependent on the damage energy, but independent of the cascade volume. They also showed that the volume of the molten zone could be greater than that of the displacement cascade.

For displacement cascades whose volume is just greater than that of the molten zone, the disruption of focussions will increase the temperature of the surrounding volume and, perhaps, the molten zone size. This will cause more interstitials to be brought back to the molten zone, which will decrease the difference between the molten zone and lattice density. Consequently, the number of vacancies left in the center of the molten zone will decrease and the molten zone lifetime will increase. These effects will have the greatest impact on small loops and will produce more loops with a size below the microscope resolution limit. Because of this shift in size of the loops, the defect yield will decrease. For displacement cascades whose volume is significantly greater than that of the molten zone, the interstitials trapped throughout the cascade volume by the solute atoms are removed from the molten zone. The increase in the molten zone lifetime that occurs because of the change in the temperature of the surrounding material will increase the number of vacancies left in the center of the

molten zone, which will form larger loops.

The experimental observations can be understood as follows: The initial increase in the defect yield with the addition of solute is due to solute elements disrupting focussons, which increases the temperature of the material surrounding the molten zone. This results in an increase in the molten zone lifetime and a concomitant increase in the loop formation probability. If this mechanism continues to dominate, the defect yield would increase rather than decrease with increasing solute content. The decrease in the defect yield as the solute level is increased is due to a combination of solute effects: (a) the density difference between the molten zone and the matrix is reduced because interstitials produced nearer the molten zone by the disruption of replacement collision sequences are now included in the molten zone and (b) the temperature of the lattice surrounding the molten zone is increased because of the disruption of focussons. This combination of effects has the greatest impact on displacement cascades whose volume is similar to that of the molten zone, and will reduce the number of loops close to the microscope resolution limit. This decrease in the number of small loops as the solute content increases is seen in the size distributions presented in Figures 3 and 4. The increase in the density of large loops as the solute content is increased is explained by considering the effect of focusson disruption on the molten zones in displacement cascades whose volume is large compared to that of the molten zone. Here the effect is to increase the lifetime of the molten zone and, hence, increase the number of loops left in the center at the end of the solidification process, which will increase the size of the loop.

5. Conclusions

It has been shown that the effect of increasing the level of Si or Al in Ni caused (a) the defect yield to increase and then to decrease, (b) the density of loops close to the microscope resolution limit to decrease and (c) the density and overall size of the large loops to increase. These results were explained by considering how solute element effects (i.e., disruption of focussons and replacement collision sequences and solute-interstitial trapping) changed the properties of the molten zone. These changes were then applied to the statistical distribution of cascade volumes and the consequences on the loop formation probability and size were used to explain the experimental observations.

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Figure captions.

Figure 1. Comparison of the damage structure produced by simultaneously irradiating the different materials to a dose of 6×10^{11} ions cm^{-2} at 300 K. A) Ni, B) Ni-0.6 at.% Si, C) Ni-4 at. % Si, D) Ni-0.5 at.% Al E) Ni-5 at. % Al and F) Ni- 7 at. % Al.

Figure 2. Defect yield as a function of alloying content for samples irradiated to a dose of 6×10^{11} ions cm^{-2} at 300 K.

Figure 3. Distribution of the loop image diameters in Ni and the Ni-Si alloys irradiated to a dose of 6×10^{11} ions cm^{-2} at 300 K.

Figure 4. Distribution of the loop image diameters in Ni and the Ni-Al alloys irradiated to a dose of 6×10^{11} ions cm^{-2} at 300 K.

Figure 1

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