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Radiation-Induced Segregation and Grain Boundary Migration in Fe-Cr-Ni Model Alloy under Irradiation

Heishichiro Takahashi and Naoyuki Hashimoto

Faculty of Engineering, Hokkaido University, Sapporo 060, Japan

A Fe-Cr-Ni model alloy was electron-irradiated using a high voltage electron microscopy (1000 kV), and *in-situ* observations on structural evolution and microchemical analyses were carried out. When the Fe-Cr-Ni alloy was irradiated, the nucleations of dislocation loops followed by voids were observed and at the same time when a grain boundary migration occurred. The compositional analysis after irradiation of an area including a grain boundary indicated nickel enrichment and chromium depletion near the grain boundary. It is suggested that when the radiation-induced point defects flow into the grain boundary, boundary migration and solutes redistribution are induced and the magnitudes depend on net point defects flow, especially that of interstitial atoms.

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Keywords: *segregation, irradiation, point defects, sink boundary migration, recombination*

I. Introduction

Austenitic stainless steels, such as type 316, have been considered as the primary candidate structural materials for fusion reactors, and are used as light-water reactor core components[1]-[3]. In these steels radiation-induced solute redistribution (segregation) at grain boundaries and grain boundary migration cause significant deleterious effects on their physical, chemical and mechanical properties[4]. Therefore it is of great importance to investigate the mechanisms for retardation of radiation-induced solute redistribution (RISR)[5][6] and then the behaviors of grain boundary migration under irradiation[7][8].

This paper is focused on RISR and the grain boundary migration behavior during electron irradiation of a Fe-Cr-Ni alloy. The grain boundary migration behavior is also discussed on the basis of solute segregation and point defect flow.

II. Experimental

The specimen used was Fe-15Cr-20Ni (0.003 mass%C, 20.10 mass%Ni, 15.20 mass%Cr, 0.0011 mass%N). After heat treatment at 1273 K for 30 min, the specimens were irradiated with 1 MeV electrons in the temperature range of 743 to 803 K using a high voltage electron microscope. The mean damage rate was 5.0×10^{-4} dpa/s. The chemical compositions were analyzed after irradiation using an energy dispersive X-ray analyzer equipped with a 200 kV transmission electron microscope. The specimen orientations for irradiation were mainly ⟨110⟩ and ⟨122⟩ or ⟨123⟩.

III. Results

1. Microstructural changes during electron irradiation

When the alloy was irradiated, the formation of defect clusters such as dislocation loops were formed initially in the matrix and voids were nucleated subsequently. Figure 1 shows a typical series of microstructural changes at 773 K during irradiation of a region including a grain boundary. During the irradiation, the grain boundary in the irradiation region migrated, although no grain boundary migration occurred in the un-irradiated area. Similar damage structures were observed at other irradiation temperatures. Figure 2 showed the effect of irradiation temperature on void number density after irradiation to 1, 2 and 3 dpa. The densities were measured in areas removed from grain boundaries. It is obvious that the void number densities at each dose showed a minimum value between 763 and 783 K. This result indicates that the void nucleation was remarkably retarded between 763 and 783 K. This retardation of void formation may be related to the decrease of super-saturated vacancy concentration as a result of preferential defect flow toward the grain boundary sink and or the enhancement of mutual defects recombination.

2. Grain boundary migration during irradiation

The grain boundary migrations were observed during irradiation at 763 to 783 K, and the occurrence of boundary migration seems to be dependent on the orientation relationship of boundary interfaces. Namely the migration did not occur when the crystalline orientations of the interface between two grains are close and the grain boundary consisting of two grains with a large orientation difference such as ⟨110⟩ and ⟨122⟩ or ⟨123⟩

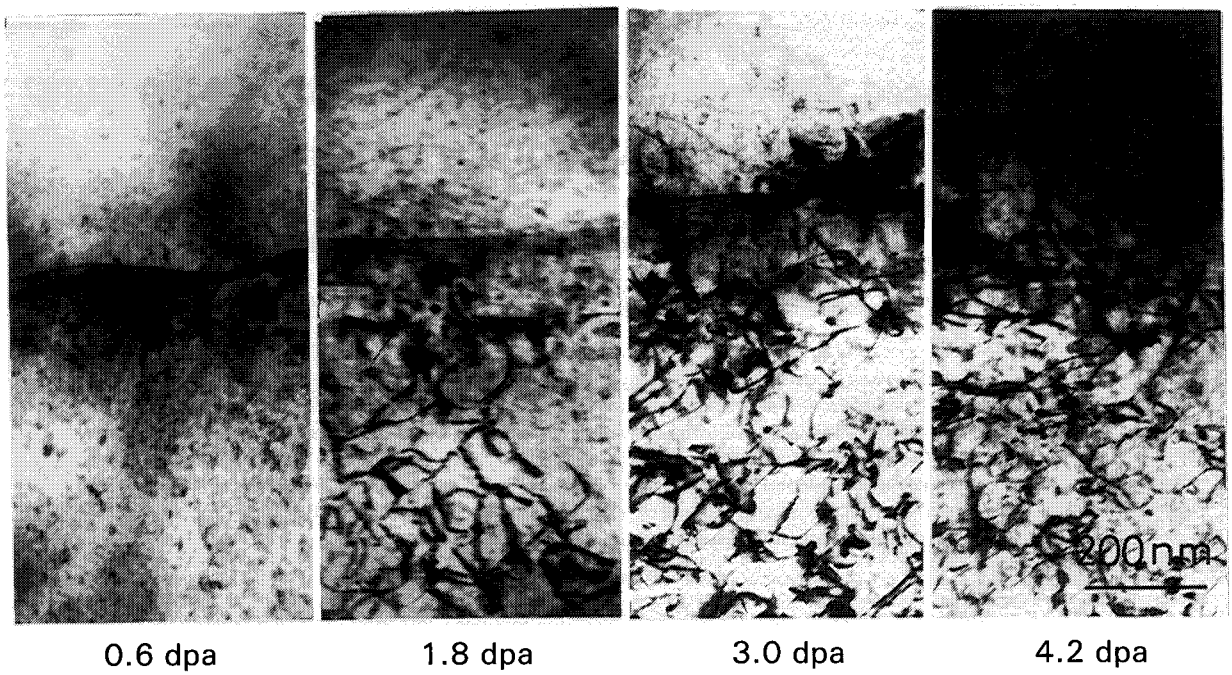

Fig. 1 Microstructural changes and grain boundary migration at 723 K.

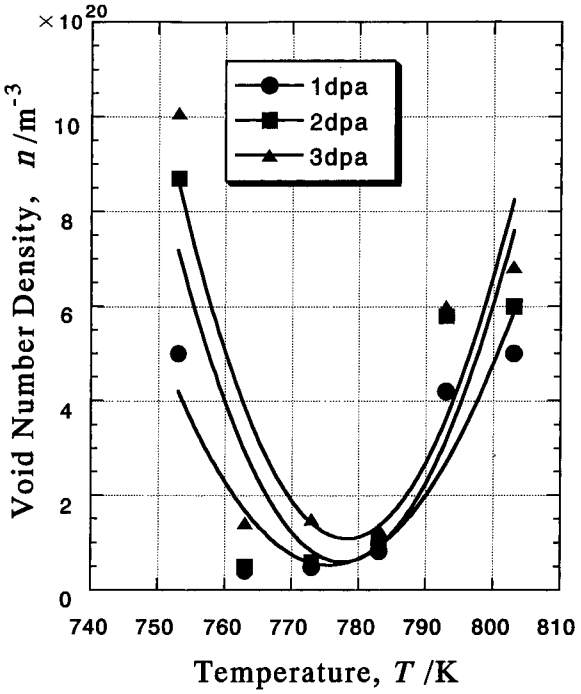

Fig. 2 Void number density as a function of irradiation temperatures after irradiation to 1, 2 and 3 dpa.

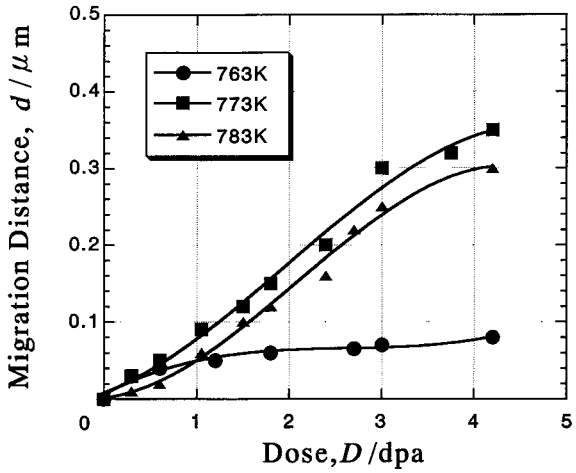

Fig. 3 Grain boundary migrations as a function of irradiation doses during irradiation at 763, 773 and 783 K.

migrated remarkably.

The boundary migration began before the formation of voids and continued to migrate even after the void nucleation. When the migrating boundary front reached some voids, they shrank quickly and the boundary migration velocity was accerelated.

Figure 3 showed the relation between the boundary migration and irradiation dose in the temperature range of 763 and 783 K. There is no remarkable difference in the migration distance up to 1 dpa. However, above 1 dpa, the distance strongly depended on the irradiation temperature. At a lower temperature of 763 K, the migration distance is not largely decreased even at a higher irradiation dose, but at higher temperatures of 773 and 783 K the distance increased with increasing irradiation dose. The migration rate at the two temperatures was about 0.04 nm/s. The distances of boundary migration at given doses of 1.8 and 4.2 dpa are shown in Fig. 4 as a function of irradiation temperature. A peak value of the migration distance was obtained at 773 K irradiation. The temperature of maximum grain boundary migration is close to that of minimum void number density.

3. Solute segregation near grain boundary

After irradiation of a region including a grain bound-

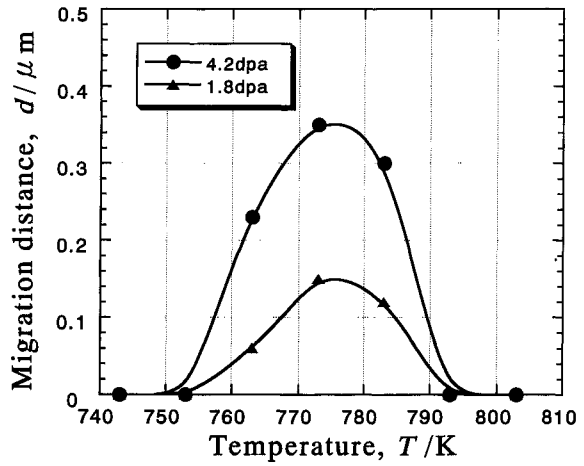

Fig. 4 Temperature dependence of grain boundary migration distance after irradiation to 1.8 and 4.2 dpa.

ary, the solutes concentration in this area was analyzed using EDS. Figure 5(a), (b) and (c) showed a typical concentration profiles of major solute atoms of nickel and chromium after irradiation to 4 dpa at 763, 773 and 783 K. The boundary positions of before and after migration were indicated in the figures and the arrow means the boundary migration direction during irradiation. As the general characteristics of the segregation behavior, nickel was enriched and chromium was depleted in the grain boundary region. Outside the area of nickel enrichment and chromium depletion, the opposite decomposition layers, namely nickel depletion and chromium enrichment, were formed. It was also recognized that, at lower temperatures, the solute concentration changes took place only in the vicinity of the last position of the migrating boundary. This is indicating that solute redistribution took place on the migrating boundary. The opposite concentration changes of chromium and nickel outside the region of the migrated boundary was larger at the lower irradiation temperatures, whereas the concentration profile in the same region tended to become flat at the higher temperatures.

Figure 6 showed the temperature dependence of the amount of solute segregation which was defined as a difference between the concentration at the grain boundaries irradiated and un-irradiated. Nickel enrichment and chromium depletion took place at all temperatures examined. Both concentration changes were prominent at 773 and 783 K.

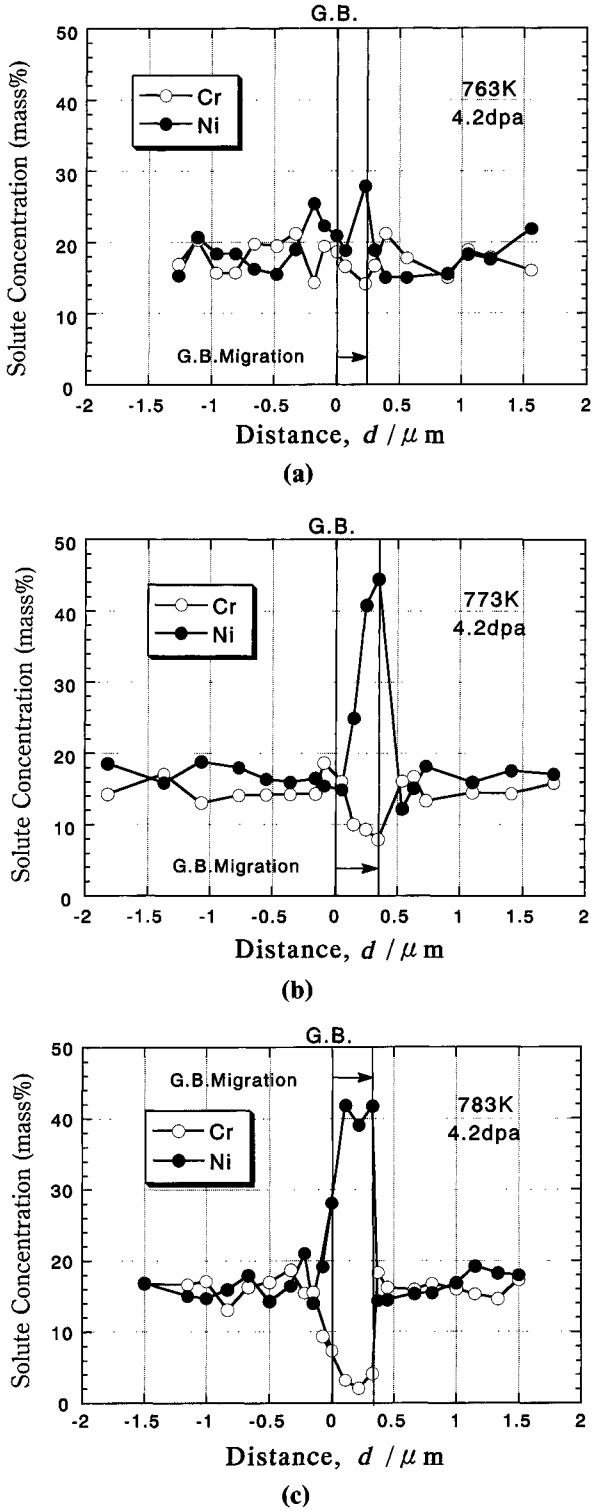

Fig. 5 Concentration profiles of Ni and Cr near a grain boundary during irradiation (a) at 763 K, (b) at 773 K and (c) at 783 K. (Initial position of the boundary was referred to as zero distance and the arrow shows the direction of boundary migration.)

IV. Discussion

As described above, the temperature dependence of void nucleation and segregation showed a similar tendency to that of grain boundary migration rate and the migrated distance. These facts suggest that the behavior of grain boundary migration should be related to that of radiation introduced point defects. That is, a coincidence of maximum changes of solutes concentration and migration distance at a given dose indicates that the point defect flows are closely associated with the grain boundary migration. The point defects of vacancies and interstitial atoms introduced during electron irradiation tend to migrate toward the point defects sinks such as the grain boundary and specimen surface. As the grain boundary is a strong defects sink in a polycrystal, the

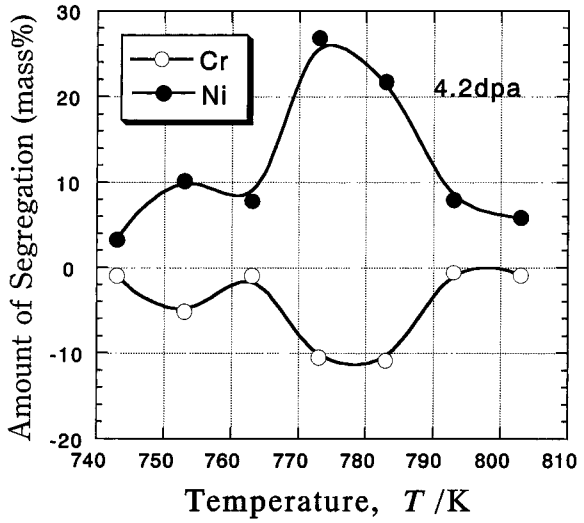

Fig. 6 Temperature dependence of the amount of solutes segregation (or depletion) after irradiation to 4.2 dpa.

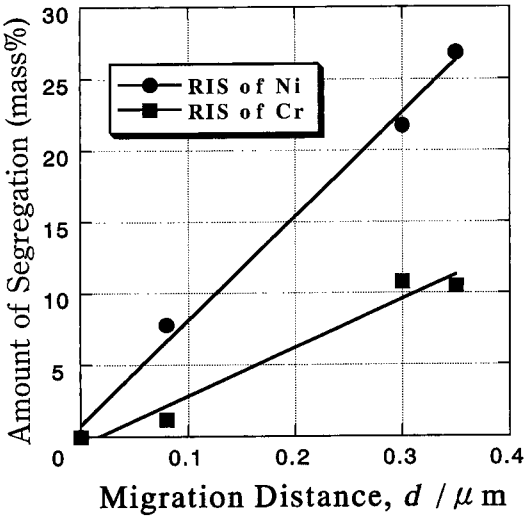

Fig. 7 Amount of solute segregation as a function of the migration distance of grain boundary. For Cr showing the amount of depletion (negative segregation) are plotted.

point defects partly flow into grain boundaries. Consequently, the super-saturated vacancy concentration is decreased in the matrix nearby grain boundary so that void nucleation is retarded. It is also suggested that segregation of solutes in materials take place due to the size effect of the solute atoms[9][10], namely over-sized chromium atoms are depleted as a result of interaction with vacancies and undersized nickel atoms are enriched according to mixed dumbbell mechanism of interstitials[11]. Therefore, the occurrence of radiation-induced segregation at grain boundaries means that the point defects were flowing into the grain boundaries during irradiation. The enrichment of nickel and depletion of chromium atoms suggest a simultaneous flow of interstitials and vacancies to the grain boundaries.

Figure 7 showed a relation between the amount of solute segregation and grain boundary migration distance. The amount of segregation increased with the migration distance. This suggests that in the process of boundary migration the solutes concentration should continue to change at the grain boundary. This tendency is remarkable for nickel rather than chromium. The diffusivity of interstitials are higher than that of vacancy. For example at 723 K, the diffusivity of interstitials was about twice higher than that of vacancy. Furthermore, the present results showed that the amount of nickel atoms moved toward the grain boundary is larger than that of chromium diffused away from the boundary. These may suggest that the role of interstitials should be more important for grain boundary migration under irradiation, though from present experiment the precise effective flux of point defects was not estimated. It is thus suggested that when the radiation-induced point defects flow into the grain boundary, boundary migration and solute redistribution are induced and the magnitudes depend on the net point defect flow. The more detailed mechanism of boundary migration related to the point defect flow with solute redistribution should be studied as a future investigation, especially by considering the nature of the boundary interface.

V. Summary

During electron irradiation of Fe-Cr-Ni steel, the redistribution of nickel and chromium solutes near the grain boundary and the grain boundary migration occurred as well as the void formation. The boundary migration was remarkable in the temperature range where the segregation of solutes was prominent. The relative amount of segregation of nickel solutes was higher than that of depletion of chromium under the same irradiation condition. From these results it is suggested that the grain boundary migration is strongly attributed to the point defects flow into the grain boundary. The migration mechanism should be investigated on the basis of the defects flow and atom rearrangement at the boundary interface.

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