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Destination of point defects and microstructural evolution under collision cascade damage

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Abstract

Microstructural evolution strongly depends on the annihilation sites of point defects and the time sequence for point defect reactions. Factors to determine the destination of point defects in collision cascade damage are categorized and the mechanism to control the destination is examined. Major factors that determine the destination are the initial distribution of point defects, the type and distribution of annihilation sites, and the style of migration of point defects and their mobility. The initial distribution of point defects varies with primary recoil energy, materials parameters and damage rate. Major annihilation sites for point defects are point defects, point defect clusters and permanent or fixed sinks such as dislocations, grain boundaries, surfaces, etc. Microstructural evolution strongly depends on the geometrical relationship between annihilation sites and cascades. The mobility and direction for migration of point defects are changed by the irradiation temperature and the strain fields from various sources, respectively. The consequence of the different mode of point defect migration is also discussed.

1. Introduction

Although equal number of interstitials and vacancies are formed in a collision cascade, microstructural evolution is different due to irradiation conditions and materials parameters. Such factors change the destination of point defects. They change the annihilation sites of point defects and the time sequence of point defect reactions. The aim of this paper is to categorize the factors that determine the destination of point defects. Most of experimental results demonstrated are for fcc metals. But the categorization is applicable to wide range of materials, if point defect reactions play an important role in the defect structural evolution. The mechanism to control the destination is examined and the contribution of each factor to the development of microstructural evolution is discussed.

Three major factors to determine the destination are considered. The first is the initial distribution of point defects. The second is the type and distribution of annihilation sites. The third is the style of migration of point defects and their mobility. Experimental results of fission neutron irradiation and fusion neutron irradiation under a variety of irradiation conditions are analyzed based on the categorization of these factors, and the advantage of such a categorization for understanding microstructural evolution is demonstrated.

2. Initial distribution of point defects in a cascade

The initial distribution of point defects varies with primary recoil energy spectrum and materials parameters. The size distribution of cascades is determined by primary recoil energy spectrum, which depends on incident particles and their energy spectrum. A comparison of the microstructures produced by fission neutron irradiation and fusion neutron irradiation is shown in Fig. 1. Thin foils of Au, which had been prepared for electron microscope observation in order to observe defect structures formed directly by cascade damage, were irradiated with 14 MeV fusion neutrons at 563 K and fission neutrons at 573 K. At these temperatures, no subcascade structures were observed. Only a large

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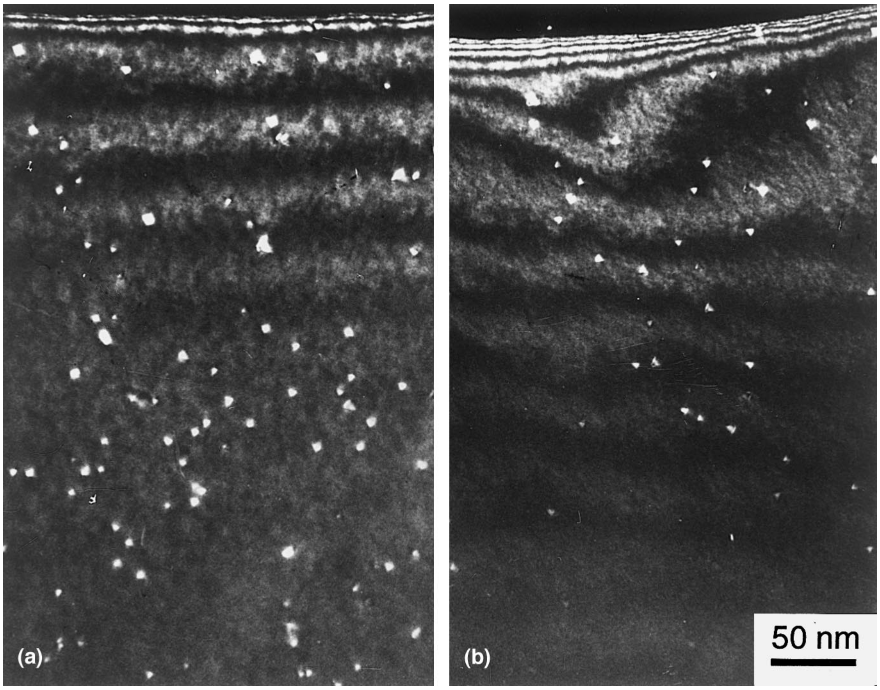

Fig. 1. Comparison of defect structures in thin foil irradiated Au between (a) 14 MeV fusion neutron irradiation, 6×10^{22} n/m^2 at 563 K and (b) fission neutron irradiation, 3.7×10^{23} n/m^2 ($E > 1$ MeV) at 573 K.

stacking fault tetrahedron was formed from a cascade. In spite of the high total irradiation fluence from fission neutron irradiation, the number density of stacking fault tetrahedra was low and the average size was small. This is caused by differences in the primary recoil energy spectrum between fission neutrons and fusion neutrons. Small cascades formed by fission neutrons cannot produce stacking fault tetrahedra. They are thermally unstable at these temperatures and evaporate during the irradiation.

Compactness or diluteness of cascades is determined by materials parameters. The difference in the size of defect clusters formed directly by cascade damage is shown in Fig. 2, where Au, Ag and Cu were irradiated as thin foils by fusion neutrons at room temperature. In this irradiation condition, each subcascade in a cascade forms stacking fault tetrahedron. The subcascade structure of Au is smaller than that of Ag and Cu because of the compactness of the cascade.

The distributions of vacancies and interstitials produced by collision cascades are expected to be different from each other. The simplest picture is that highly condensed vacancies are surrounded by interstitials. The effect originating from the difference of the initial distributions of point defects is called the Cascade Localization Induced Bias (CLIB) effect [1,2]. Vacancies require more jumps than interstitials before they annihilate at sinks outside the cascade. Therefore, the reactivity of vacancies in the cascade area is higher than that of interstitials. From this effect, vacancy clusters in a cascade absorb more vacancies than interstitials and grow larger. Strong void growth is expected when the differences in point defect distribution are large.

3. Annihilation site for point defects

Three kinds of major annihilation sites for point defects are considered. The first is mutual annihilation with the opposite types of point defects. If the cascade is a compact cascade, the concentrations of point defects in the cascade are high. Consequently, the mutual annihilation rate is high. If the primary recoil energy is high and a cascade is subdivided into well defined subcascades, the annihilation rate is also high due to inter-subcascade interactions.

The second is absorption of point defects by point defect clusters of the same or opposite types of point defects. If the clusters are in a cascade, the growth of vacancy clusters and the shrinkage of interstitial clusters occur by the CLIB effect as noted in the previous section. Under an irradiation condition where freely migrating point defects play an important role, the stability of defect clusters changes the destination of point defects. If interstitial clusters and vacancy clusters nucleate at the same time, both can grow larger, as for the case of Ni–2 at.% Ge shown in Fig. 3(a). Ni–2 at.% Ge was irradiated in bulk at 573 K by fission neutrons using the

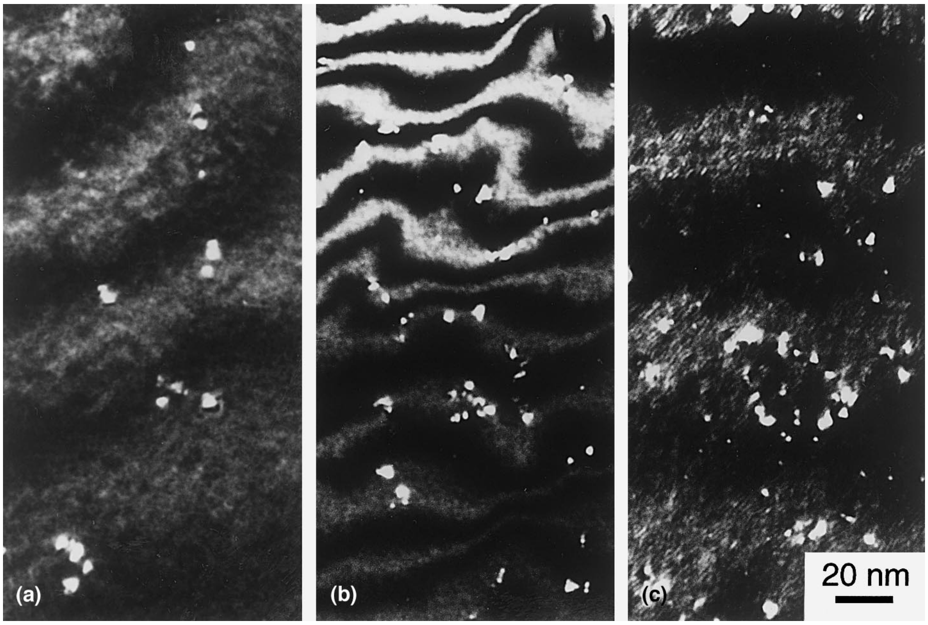

Fig. 2. Comparison of defect structures between (a) Au, 3.5×10^{20} n/m^2, (b) Ag, 2.1×10^{20} n/m^2 and (c) Cu, 1.5×10^{21} n/m^2, irradiated as thin foils with 14 MeV fusion neutrons at room temperature.

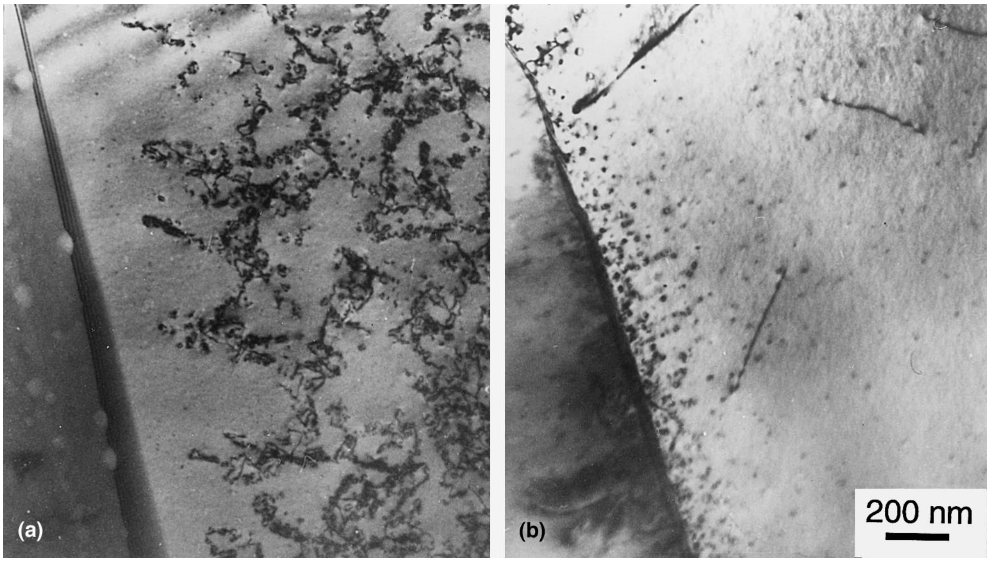

Fig. 3. Comparison of defect structures between (a) Ni–2 at.% Ge and (b) Ni–2 at.% Sn, irradiated with fission neutrons as bulk, 3.7×10^{23} n/m^2 ($E > 1$ MeV) at 573 k. Note the defect structures near grain boundaries (on the left side in Figures).

JMTR (The Japan Materials Testing Reactor). Bulk irradiation means specimens are polished for electron microscopy observation after irradiation. If only one type of cluster nucleates and grows at an irradiation condition, another type of point defect remains and prevents further growth of the clusters. An example is shown in Fig. 3(b). In this irradiation condition, only interstitial type dislocation loops can nucleate. In an area far from the grain boundary, few loops are observed.

The third kind of annihilation site is permanent or fixed sinks such as dislocations, grain boundaries, surfaces, etc. Microstructural evolution strongly depends on the geometrical relationship between permanent sinks and cascades. Neutral permanent sinks act to change the destination of freely migrating point defects and compensate the unbalanced point defect reactions. For example, for the condition where only interstitial type dislocation loops nucleate, loops can grow larger near neutral sinks such as surfaces and grain boundaries as shown in Fig. 3(a). Remaining vacancies which prevent the growth of loops are absorbed by the sink.

For an understanding of void growth, the destination of interstitials should be considered. In the dislocation bias theory, the absorption of interstitials by edge dislocations is essential for the accumulation of vacancies in voids. For void growth by the CLIB effect, the destination of vacancies is determined by an unbalanced reaction between interstitials and vacancies in a cascade. Sinks are required to absorb the remaining interstitials. But in this case, no bias for sinks is essential. This point is different from the dislocation bias theory. In the recent production bias theory [3,4], interstitials are absorbed by sinks before accumulation of vacancies. In this case, the bias of sinks is also not significant.

4. Mobility and migration of point defects

The point defect development strongly depends on the mobility and direction of migration of point defects. The mobility determines the absolute speed of point defect reactions and also determines the point defect concentration. If the mobility is high, remarkable defect structural developments are expected. The difference in the mobility of two types of point defects causes a time lag for point defect reactions [5]. At first, interstitials are released from a cascade. If the difference of the mobility is high, interstitial reactions are superior to vacancy reactions. Pre-existing interstitial clusters are favored to grow, while vacancy clusters may be annihilated by absorption of interstitials. This condition is only expected when the irradiation dose rate is low enough. When the irradiation rate is high, cascade formation rate is also high. Vacancies can arrive at pre-existing interstitial clusters at the same time as interstitials, and the effect of the time lag is not significant.

The stress field at edge dislocations determines the direction of point defect flow. Interstitial type dislocation loops are formed in the area which has a dilatational field at edge dislocations. These loops are observed in the initial stage of irradiation as shown in Fig. 4, where the nucleation of loops is not easy in the matrix. Embryos of interstitial type dislocation loops are formed directly by the cascade damage. They can grow preferentially by excess flow of interstitials to the dilatational area at edge dislocations. With increasing irradiation fluence, the growth of loops in the matrix is observed as shown in Fig. 4(d).

The consequence of the different mode of point defect migration should be considered. Recently one dimensional motion of interstitial clusters which are formed in the cascade have been reported as one type of point defect migration. They are considered to be bundles of crowdions and move in ⟨1 1 0⟩ directions. One jump length for moving clusters is longer and the reactivity of clusters is weaker than for the usual random walk of interstitials when the diffusion is in the same direction. Consequently, the mutual annihilation rate of moving clusters is low, which contributes to defect structural development. The moving clusters also contribute to the preferential growth of loops at one side of edge dislocations by their one dimensional motion by the stress field of dislocations and loops as shown in Fig. 4.

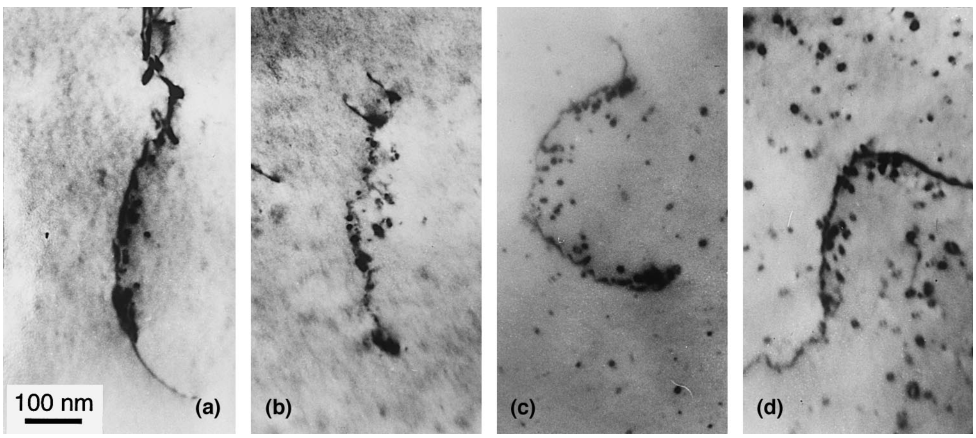

Fig. 4. Interstitial type dislocation loops formed near edge dislocations in Ni irradiated with 14 MeV fusion neutrons as bulk, (a) 7.8×10^{20} n/m^2, (b) 1.4×10^{21} n/m^2, (c) 6.0×10^{21} n/m^2 and (d) 6.0×10^{22} n/m^2 at 563 k.

5. Advantage of the categorization

5.1. Temperature

Irradiation temperature is one of the most important factors to determine defect structural development. It changes the destination of point defects remarkably. At lower temperatures, point defect clusters are formed directly in a cascade as intra-cascade reactions which decrease the total amount of freely migrating point defects and consequently significant defect structural development is not expected. At higher temperatures, the nucleation of clusters in cascades is not easy and most of point defects migrate as freely migrating point defects. In this condition, their main destination is point defects and point defect clusters.

The mobility of point defects strongly depend on the irradiation temperature. It changes not only the absolute speed of point defect reactions but also the ratio of the mobility of two types of point defects. The latter causes the time lag for point defect reactions.

Recently irradiations in which temperature was intentionally changed, temperature variation irradiation, were performed in order to investigate the point defect processes [6–8]. The effect of temperature variation is understood as it varies the destination of point defects by changing point defect mobility and stability of point defect clusters. Vacancy clusters formed at low temperatures become unstable and evaporate at high temperatures. Vacancies released from the clusters and newly formed point defects which have higher mobility promote annihilation of interstitial type dislocation loops.

5.2. Alloying elements

The addition of alloying elements changes defect structural evolution remarkably. Alloying elements block the replacement sequence collision, which causes differences in the distributions of interstitials and vacancies to be small. Consequently the destination for the mutual annihilation increases. Alloying elements also change the mobility of point defects, as over-size elements and under-size elements react with interstitials and vacancies, respectively. The decrease in mobility of one type of point defect will lead to the time lag in point defect reactions so that for both types of point defects, a similar effect at low temperature irradiation will occur.

5.3. Damage rate

The damage rate changes the cascade size due to overlap of defect reactions between cascades. If the damage rate is high enough as is the case for fusion irradiation, small cascades will fuse into one and act like larger cascade damage. When embryos of defect clusters are unstable, the time lag for point defect reactions is important. If damage rate is low and the evaporation of embryos occurs before their growth, point defect clusters do not become the destination for point defects. Consequently, the mutual annihilation rate increases and cluster growth is suppressed.

Usually, defect structures in collision cascade damage are analyzed by solving the kinetic equations. The simple categorization by destination of point defects will give us a strong insight into defect structural evolution and contribute to materials design and development.

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