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Progress of Research on the Mechanism of Microstructure Evolution during Irradiation with Collision Cascades

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Progress of the research on microstructural evolution during radiation damage accompanied with collision cascades, particularly by neutrons, is summarised from the researches reported in 1995 and 1996. The major part of irradiation was performed with JMTR (Japan Material Testing Reactor), and comparisons were made with the result of D-T fusion neutron irradiation with RTNS-II (Rotating Target Neutron Source, LLNL). The subjects concerned are: (1) Microstructure evolution during irradiation, (2) Processes controlling the accumulation of defects during cascade damage, (3) Subcascades from the viewpoint of point defect cluster formation, (4) Mechanism of suppression of microstructure evolution during temperature cycle neutron irradiation of nickel, (5) Identification of the nature of neutron-irradiation-induced point defect clusters in copper by means of electron irradiation, (6) Easy one-dimensional motion of small interstitial clusters, (7) Identification of the nature of small point defect clusters in neutron irradiated Fe-16Ni-15Cr, (8) Spatial distribution of nucleation of point defect clusters by high energy particle irradiation, (9) Crystallographic orientation dependence of dislocation structures in neutron irradiated metals, (10) Examination of defect accumulation mode with multi-section-removable-irradiation-rig in Japan Material Testing Reactor, (11) Estimation of freely migrating point defects during cascade damage by the growth of helical dislocations, (12) Identification of the nature of neutron-irradiation-induced small point defect clusters in Nickel by means of electron irradiation, (13) Thermal stability of point defect clusters in neutron irradiated Fe-16Ni-15Cr.

KEYWORDS: neutron radiation damage, fusion reactor materials, fusion neutron, reactor irrdiation, collision cascade, microstructure, interstitial atom, lattice vacancy

1. Microstructure Evolution during Irradiation[1)]

Microstructural evolution in metals and alloys during energetic particle irradiation is reviewed with emphasis placed on the underlying defect reaction processes. The microstructures produced by electron irradiation with a high-voltage electron microscope, fusion-neutron irradiation with a D-T fusion neutron source, fission-neutron irradiation with reactors and ion irradiation with accelerators are compared and contrasted. The section of the damage produced by electron irradiation starts with a description of the development of the defect structure due to primary damage, and then extends to include the measurement of point defect properties, and the detection of a variety of point defect processes such as radiation-induced diffusion, the stochastic fluctuation of point defect reaction, and the instability of small interstitial clusters. The section on neutron irradiation begins with the detection and analysis of defects produced by cascades and subcascades and leads to an analysis of the recoil energy spectrum. The accumulation mode of defects is categorized in terms of the competition between defect formation by cascades and the interaction of these defects with freely migrating point defects. A direct comparison is made of the damage produced by fission- and fusion-neutron irradiation, to find a difference of more than 10 times depending on the defect processes involved. A wide variation of defects produced by mono-energetic recoil is detected by heavy-ion irradiation and the role of freely migrating interstitials is examined as a function of permanent sink strength.

2. Processes Controlling the Accumulation of Defects during Cascade Damage[2)]

All the experimental information from irradiations with fusion and fission neutrons, light and heavy ions, and high energy electrons is incorporated to establish the understanding of the mechanism of defect accumulation under cascade damage.

Five stages of sequential variation of defect accumulation modes have been extracted from experiments. Key component processes are the competition and cooperation of the direct formation of defects from cascades and the reaction of freely migrating point defects. 1st stage of very low dose to disclose vacancy clusters from cascades by the impact effect from succeeding ones, 2nd stage of simple accumulation vacancy clusters by the prompt escape of interstitial atoms, a modification of the 2nd stage of the accumulation of vacancy clusters along with the formation of interstitial clusters nucleated at cascades and grown by freely migrating interstitials, the 4th stage of the saturation by the geometrical overlap of cascade zones.

Brief explanations of related experiment s and analyses will be included. Identification of the nature of small point defect clusters produced by collision cascades in a variety of materials, from their behavior during additional electron irradiation. Primary recoil energy spectrum effect from the comparison of irradiations with a wide variety of energetic particles. An important role of stochastic fluctuation of point defect reaction in determining the success probability of the nucleation of point defect clusters. One dimensional easy motion of small interstitial clusters to remove interstitials from cascade damage zone and to determine the crystallographic orientation dependence of dislocation structures. A characteristic behavior of cascade produced point defect clusters during irradiation with cyclic temperature variation.

3. Subcascades from the Viewpoint of Point Defect Cluster Formation[3)]

Splitting of large collision cascades into separated subcascdes is well defined in medium-weight materials (Cu and Ni). In light-weight materials (Al), all the collisions are not closely spaced and subcascades cannot be defined. Toward the other direction to heavier-weight materials (Ag and Au), collisions are densely packed and separation into subcascades is generally not considered important. The aim of this paper is to reconsider the concept of subcascades in heavy-weight metals from the viewpoint of the formation of point defect clusters by means of Binary-Collision-Approximation simulation.

The first part deals with the variation of the spatial spread and localization of collisions started from primary knock-on atom (PKA) of the same energy and random direction. For example, 200 keV PKA produced always approximately 4000 displaced atoms, but the degree of the localization of displaced atoms were widely different: they are formed within a small volume of 10000 or they are widely expanded to a large volume of 100000 atoms for the other extreme case. Towards the latter case of extreme, the number of separated localized volume of high collision density increased.

The second part deals with the variation of collision distribution by a collision density criterion. When an appropriated high value is assumed for a threshold collision density, several localized volumes are extracted in comparatively wide spread collision zones, for example ;up to 10 for the case of 200 keV PKA. When the threshold density is assumed below certain value, all the collision zones is united to a continuous volume.

The consequence of the wide variation from the same energy PKA appeared as the wide variation of vacancy cluster groups in the mono-energetic heavy-ion irradiation. The consequence of the variation with collision density criterion appeared as the temperature dependence, a number of closely spaced small vacancy clusters at vacancy immobile low temperatures and a single large cluster at vacancy mobile high temperatures. Here an important remark is made; the splitting of a large collision cascade into subcascades should no be uniquely defined but depend on the kind of reactions after the collision in interest.

4. Mechanism of Suppression of Microstructure Evolution during Temperature Cycle Neutron Irradiation of Nicke [4)]

In nickel irradiated with cyclic temperature variation between low and high temperature in a reactor JMTR, the interstitial loops was much less than those expected from the lower temperature irradiation. Furthermore, they were less than in specimens irradiated at the higher temperature of the combination. This strong suppression of interstitial type defect structure development has been suggested to be due to the reaction of high density of small vacancy clusters produced by the lower temperature irradiation (M. Kiritani et. at.: J. Nucl. Mater. 212-215 (1994) 241-245). The purpose of this paper is to clarify the mechanism of suppression of dislocation structure evolution during temperature cycle neutron irradiation.

Since the annealing-out mechanism of small vacancy clusters in the form of stacking fault tetrahedra (SFT) which are the defects produced in nickel by lower temperature neutron irradiation is not well known, their thermal stability data were obtained experimentally from the annealing experiment of electron-irradiation induced SFT. Upon annealing, SFT shrink and disappear indicating the emission of vacancies, smaller ones at lower temperatures. Applying the annealing data to SFT produced by one cycle of lower temperature neutron irradiation at 473 K, taking account of SFT size distribution, 30 % and 80 % are expected to disappear by 30 and 90 min respectively after the rise of temperature to 673 K. The concentration of vacancies released during this annealing is estimated to be about $3x10^{-5}$, and this is far enough to eliminate coexisting interstitial clusters which contain $7x10^{-6}$ interstitials. Thus the interstitial loops nucleated during irradiation at lower temperatures cannot serve as the origin of dislocation at higher temperature.

On the other hand, about 10 % of SFT, larger ones in size, can survive the higher temperature irradiation, and they prevent the formation of new interstitial clusters by absorbing freely migrating interstitials.

Vacancy clusters introduced during irradiation at lower temperatures are playing two roles: first is the elimination of lower-temperature-irradiation-induced interstitial clusters by evaporating vacancies during an early stage of higher temperature irradiation, and the second is the suppression of the nucleation of new interstitial clusters during higher temperature irradiation. Simulation and analysis of the sequential reaction are also reported.

5. Identification of the Nature of Neutron-Irradiation-Induced Point Defect Clusters in Copper by Means of Electron Irradiation[5)]

Identification of the nature of small point defect clusters is prerequisite for the progress of the investigation of defect microstructure evolution by neutron irradiation. A powerful new technique was introduced in which the identification is made from the behavior of defects under electron irradiation in a high voltage electron microscope. This technique is especially effective for small clusters such as those produced directly from collision cascades. At the start of an electron irradiation during which the reaction of interstitials dominates that of vacancies, vacancy clusters are expected to shrink and disappear, and interstitial clusters are expected to grow.

Point defect clusters examined were those produced in copper thin foils and bulks by three irradiation facilities: fusion neutron source RTNS-II (mono-energetic 14 MeV), fission reactor JMTR (up to 2 MeV) and high energy neutron source LASREF (up to 100 MeV). The electron irradiation was performed with 1 MeV electrons at 300, 363 and 473 K.

Large percentage of small defect clusters in samples neutron-irradiated as both thin foils and bulk specimens disappeared in a few minutes, so they were judged to be vacancy type. In detail, about 95 % of the clusters in thin foils were identified to be of vacancy type. In bulk specimens some of the larger clusters grew, and a presence of interstitial-type defect clusters is confirmed. In the case of bulk specimens, variation with the neutron irradiation facility is observed: the proportion of interstitial type clusters introduced by JMTR irradiation was less than by RTNS-II and LASREF.

These results of the identification by electron irradiation agreed with our former judgment by electron microscopic image contrast, with proper selection of diffraction condition in bright and dark field imaging.

In a very thin part of a specimen, some of vacancy type clusters with SFT contrast were observed to grow during electron irradiation at room temperature, and this can be understood by an extra-accumulation of vacancies as the result of the faster escape of interstitials to surfaces.

6. Easy One-Dimensional Motion of Small Interstitial Clusters[6)]

Taking advantage of extremely high damage rate in electron irradiation with a high voltage electron microscope (>10^4 times of a strong reactor), characteristics of the motion behavior of small interstitial clusters have been extracted. (1) Crystallographic direction of the motion is along closely packed direction, along [110] in fcc and along [111] in bcc metals that have a large angle to the plane of loop. (2) Distance of the motion is comparable to the separation of interstitial clusters. (3) Speed of the motion is faster than detected by normal video-recording. (4) The clusters often exhibit a quick back-and-forth motion between two positions. (5) Correlated motion is often observed, in which clusters move sequentially after the motion of other near-by clusters. (6) A large number density of clusters is required for the motion so that the spacing between clusters is not far more than the size of clusters. (7) A favorable circumstance for the motion of a cluster is a continuous change of the size and configuration of the surrounding clusters. (8) Electron-microscope image of a cluster which is understood from a dislocation loop becomes faint before the cluster start its motion and recovers soon after the stop of the motion.

All the observation mentioned above leads to a proposal of the motion mechanism of small interstitial clusters. The motion cannot be expected from the motion of dislocation, in which a loop moves as a whole by the gradient of shear stress as the driving force. The displacement of atoms around a cluster, at least during their motion, is thought to be extended along the motion, like a bundle of crowdion. Here, a proper name will be "bundle crowdion".

The transport of interstitials by bundle crowdion mode is expected to have important consequences in the defect structure development during neutron irradiation. It governs the success probability of interstitial cluster formation from collision cascades, and also decides the efficiency of bias effects intrinsic to collision cascades, such as production bias and cascade-localization-induced bias. It serves as the origin of crystallographic orientation anisotropy in the dislocation structures as reported in a separate paper in this conference.

7. Identification of the Nature of Small Point Defect Clusters in Neutron Irradiated Fe-16Ni-15Cr[7)]

Since size of point defect clusters produced directly from collision cascades in stainless steels are generally smaller than that in simple fcc metals, conventional methods for the nature identification utilizing electron microscope images are unreliable. We propose a new method to identify the nature of very small defect clusters from the behavior in a interstitial-dominant circumstance during a short time after the start of electron irradiation. The stereo micrographs before and after the irradiation was combined with the in-situ observation with a high voltage electron microscope.

In this paper, the results of identification of the nature of small defect clusters in Fe-16Ni-15Cr neutron irradiated with JMTR at 353-623 K are reported. Discussion is made on the process of microstructure evolution by neutron irradiation.

The defect clusters that have a similar contrast to that of stacking fault tetrahedra were identified to be of vacancy type. Fraction of them decrease with an increase of neutron irradiation temperature in the range 353-623 K, which suggest that the vacancy clusters formed directly from subcascades become unstable for higher temperature. On the other hand, interstitial clusters which nucleate at cascades grow even at higher temperatures by the association of freely migrating interstitials.

In specimen neutron-irradiated at 573 K as a thin foil, the defect structure varied remarkable with the thickness of the specimen. Identification by electron irradiation disclosed a larger fraction of interstitial type defect clusters at thicker part.

The present results have made possible to identify small defect clusters here after, utilizing the shape and contrast of defect clusters under suitable diffraction conditions of bright and dark field imagine.

8. Spatial Distribution of Nucleation of Point Defect Clusters by High Energy Particle Irradiation[8, 9]

Spatial distribution of point defect clusters in high energy particle irradiation reflect their formation mechanism. They were examined by means of a statistical method for the cases of electron, ion and neutron irradiation.

Characteristics of distribution of clusters can be examined by the analysis of the deviation from random distribution. One direction of the deviation is towards homogeneous distribution in which each cluster t ends to be separated from others. Here, 'randomness' and 'homogeness' have not been distinguished each other so far. The opposite direction of the deviation is heterogeneous distribution in which each cluster tends to gather in groups.

The deviations from random distributions were examined as follows. A large area for the measurement was divided into equalized blocks, and the numbers of blocks containing each number of clusters were adopted as the data to be analyzed. A random distribution is expected to obey a binomial distribution . And the standard deviation of the distribution was employed as a parameter. In homogeneous distribution, this standard deviation is smaller. In heterogeneous distribution, the deviation is oppositely larger than that in binomial distribution. In addition, these results with the standard deviations were ascertained using tests of goodness of fit for each distribution with chi-square distribution.

First, the method was applied to a simple case of electron irradiation n which each collision of incident particles produces a single Frenkel pair. A large number of interstitial type dislocation loops is generated in copper soon after the start of irradiation. The distribution of these loops were not quite random but rather homogeneous. With the lapse of time, decreasing the numbers, they approached to random distribution. Suppression of the nucleation near an existing loop is the origin of the homogeneous distribution, and random annihilation of clusters by random mobile vacancies bring to the random distribution.

In neutron irradiated gold, the distribution of stacking fault tetrahedra was far from random and homogeness. When these clusters are grouped with circles of an appropriate size, the distribution of groups obey random. It is considered that each group corresponds to a random cascade collision and each cluster to a subcascade. In addition, the size of the circle is considered to represent the mean size of a cascade.

9. Crystallographic Orientation Dependence of Dislocation Structures in Neutron Irradiated Metals[10)]

Neutron irradiation of thin foil samples has substantially contributed in extracting the defect structures directly produced from collision cascades by avoiding the reaction of freely migrating point defects. The thin foil irradiation is now serving its new role in disclosing the consequence of easy one-dimensional motion of small interstitial clusters.

In neutron irradiated polycrystalline samples, defect microstructures introduced by irradiation varied largely from a grain to grain. Typically, high number density of interstitial-type dislocation loops were developed in a grain though only voids were observed in an adjacent one. The detailed analysis of the crystallographic orientation relationship with respect to the foil specimen surface lead to the following characterization.

(1) Interstitial-type dislocation structures are less developed in samples which have all [110] directions intersect with the surface with large angles. All the small interstitial clusters produced at collision cascades is thought to have escaped to specimen surface before growing to stable loops. (2) The other way around, the majority of survived loops

have their easy motion direction parallel to the surface. (3) The fate of interstitial clusters, survival or elimination, was confirmed to be determined at a very early stage of their formation, by the examination of the orientation of very small loops after an additional growth by electron irradiation with a high voltage electron microscope. (4) Both a sequential variation along the increase of foil thickness and a variety of aligned dislocation loops related to the direction of the edge of a wedge shape specimen are results of the easy motion of interstitial clusters.

Major experimental data are for nickel irradiated in a fission reactor JMTR, and the characteristic phenomena are common to other metals such as copper and austenitic stainless steel. The details of easy one-dimensional motion of small interstitial clusters, named as 'bundle crowdion', are reported in a separate paper in this conference.

10. Examination of Defect Accumulation Mode with Multi-Section-Removable-Irradiation-Rig in Japan Material Testing Reactor[11)]

After a long laborious effort, generally only one data point to irradiation dose is obtained in reactor neutron irradiation. For example (M. Kiritani et. al.: J. Nucl. Mater. 174 (1990) 327), during the examination of fission-fusion correlation by a direct comparison of microstructural evolution, the authors were forced to be satisfied with the analysis with an assumption that the accumulation mode during fission-reactor irradiation is the same as that during fusion neutron irradiation, only in the latter the accumulation mode is known at present from the systematic irradiation.

In order to obtain information on the progress of damage structure evolution under a consistent irradiation field with a good temperature control, a new method has been proposed for the irradiation in Japan Material Testing Reactor (JMTR), in which the sample can be removed from the reactor core one after the others during one cycle of the reactor operation. Vacuum-sealed mini-capsules containing 1,000 disk specimens in each were embedded in a good thermal conductive metal block with a electric-heater for temperature control, and were removed one-by-one during one-cycle of 30 days operation. Named as multi-section-removable-irradiation-rig.

Vacancy clustered defects produced directly from collision cascades in Au, Cu, Ni and model austenitic stainless steel irradiated s thin foils at 473 K were found to increase proportionally to the irradiation dose for the range of $6x10^{20}$-$3x10^{23}$n/m2. These results justified the former analysis of fission-fusion correlation. Results on the samples irradiated as bulk including other materials will also be reported.

11. Estimation of Freely Migrating Point Defects During Cascade Damage by the Growth of Helical Dislocations[12)]

Defect structure development from collision cascades is governed by the formation of point defect clusters directly from cascades and the reaction of point defects escaped from their own cascade and migrate freely in the matrix. At present there is no standard method to measure the concentration of freely migrating vacancies and interstitials during irradiation. In this paper the amount of freely migrating point defects formed during neutron irradiation at elevated temperatures was estimated from the analysis of the growth of helical dislocations.

High energy neutron irradiation was performed for Ni at 563 k by 14 MeV D-T fusion neutrons with a rotating target neutron source (RTNS-II) and at 573 K in a fission-reactor (JMTR). After irradiation each specimen was thinned and the diameter of helixes was measured as a function of irradiation intensity using an electron microscope.

The diameter of helixes increased proportionally with the irradiation intensity for the same irradiation time of D-T neutron irradiation. This proportional increase with intensity indicates that the major annihilation process of point defects is not the mutual annihilation but the simple annihilation at permanent sinks. Further analysis showed that the amount of freely migrating point defects is less than 0.04 of NRT model estimation when 50 % of the bias factor is assumed.

The diameter of interstitial type dislocation loops formed in the same samples one order of magnitude smaller than the turn-diameter of helixes. The reason for the difference might he in the unknown birth time of each loop and also in much smaller bias factor for faulted loops. The proposed method using the growth of helical dislocation can estimate the difference in the absorption of point defects without an ambiguity in the nucleation of loops and also the difference of the bias factor for different type of dislocations.

12. Identification of the Nature of Neutron-Irradiation-Induced Small Point Defect Clusters in Nickel by means of electron Irradiation[13, 14)]

Reaction of point defects during electron irradiation are well-understood, and it is possible to identify the nature of existing defects from their behavior under electron irradiation. The major aim is to identify the nature of neutron-irradiation-induced small point defect clusters which are difficult to be determined from their electron microscope image contrasts and shapes.

When the defects are located at positions not very close to sinks such as surfaces, dislocations loops of interstitial type is expected to grow and stacking fault tetrahedra of vacancy type is expected to shrink, because strain field of those defects attract and absorbs more interstitials and vacancies.

In-situ observation during electron irradiation with a high voltage electron microscope and the comparison of the defect microstructures with stereo-microscope technique before and after electron irradiation gave us information concerning to the nature of neutron-irradiation-induced defects as follows: (1) The present method described above was proved to be efficient in identifying the nature of point defect clusters. (2) Majority of very small point defect clusters in neutron irradiated thin foil nickel are identified as vacancy type. (3) More than 3/4 of all point defect clusters in nickel induced by bulk irradiation at room temperature are clarified to be vacancy type. (4) In thin foil specimens, the fraction of interstitial clusters increased gradually with an increase of the foil thickness. Spatial distributions of both types of defect clusters in the foil disclosed the role of freely migrating interstitials. (5) Attention is necessary for the identification of defects near dislocations, specimen surfaces and grain boundaries, because they have bias effect interstitials. (6) It is confirmed that the conventional method of identification of the nature of neutron-irradiation-induced larger defects is reliable.

13. Thermal Stability of Point Defect Clusters in Neutron Irradiated Fe-16Ni-15Cr[15)]

Annealing behavior of point defect clusters in neutron irradiated Fe-16Ni-15Cr alloy with a fission reactor was examined. From the comparison of the variation of the size distribution of vacancy (V)-type clusters during annealing with the change of their size distribution by irradiation temperature, it was deduced that at higher irradiation temperature the nuclei of clusters formed directly from collision cascades do not annihilate after their formation but they do not form from the beginning. From the results of the thermal stability of defect clusters, the process of defect structure formation in a fission reactor (T-cycle irradiation), was discussed. Characteristic microstructure of T-cycle irradiation, the suppression of I-type cluster formation, was found to depend on the decomposition rate of V-type clusters accumulated at the low temperature of T-cycle irradiation after the shift to high temperature of T-cycle irradiation. When the low temperature of T-cycle radiation is lower and its high temperature is higher, the characteristic of T-cycle irradiation is enhanced.

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