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A review of in situ observation of defect production with energetic heavy ions

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Abstract

In situ transmission electron microscopy (TEM) observation of radiation damage with energetic heavy ions has been a powerful tool for the study of radiation effects since it provides means to observe evolution of cascade damage. The objective of the present paper is to summarize the status of this experimental technique, its major achievement and current problems and to suggest the future directions. Defect accumulation by irradiation with energetic heavy particles is quite different from that with electron irradiation. Spatial and temporal fluctuations occur in defect production and annihilation. These observations will be summarized together with the dependence of damage evolution on irradiation temperature, target materials, incident ions and specimen geometry. Self ion irradiation is particularly useful for the study of cascade damage. Successful results of self ion irradiation for ion–neutron correlation will be presented. The experimental issues in such in situ heavy ion radiation damage studies are that the samples have to be thin enough to be transparent to the TEM electrons and that the range of self ions to simulate a primary knock-on atom (PKA) is generally short. Moreover, damage distribution is strongly depth-dependent. We have been purposely utilizing 'thin foils', taking advantage of the strong sink effect of surfaces for mobile point defects. The issues will be discussed in terms of the potential use of this technique to derive cascade damage characteristics in the bulk. Additional techniques such as computer simulation of cascade should be incorporated in conjunction with the experiment in order to develop models of defect production, annihilation and evolution. Prospect of the experimental technique will be discussed. © 1997 Elsevier Science B.V.

1. Introduction

Radiation damage is one of the most important problems for structural materials of advanced nuclear energy systems as fission power reactors or fusion reactors. In these systems, radiation damage is created by energetic recoils from fast neutrons. For fission reactors, typical energy of a primary knock-on atom (PKA) by 1 MeV neutrons is around several tens of keV for medium-heavy target atoms as iron, whereas for D–T fusion reactors, the PKA energy is one order of magnitude higher and sometimes the maximum PKA energy will reach about 1 MeV. Such high energy PKAs will produce cascade damage, which is substantially different from simple homogeneous Frenkel pairs as produced by electron irradiation which is largely documented. Therefore, precise knowledge of the cascade is required for analyzing and estimating radiation effects on materials for the advanced nuclear energy systems.

The cascade created by high energy PKAs may be simulated by the damage created by energetic self ions. On this assumption, we have been performing in situ observation experiments of radiation damage with energetic heavy ions. The objective of the present paper is to summarize the status of the experimental technique, to discuss its major achievements and current problems and to suggest the possible directions of future investigation and their prospect.

In situ observation of radiation damage has been a powerful tool for the study of radiation effects since it provides means to observe the evolution of defects during irradiation. However, most of the efforts so far have been those utilizing high voltage electron microscopes, hence by electron irradiation which produces simple Frenkel pairs. In situ observation of cascade damage by ion irradiations

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has been studied for the past twenty years with greater experimental complexities than by the electron irradiation. Nevertheless, it has become more and more evident that the unique feature of radiation damage created by energetic heavy particles, which is substantially different from that created by energetic electrons has made us to convince ourselves the importance of in situ observation of radiation damage with heavy ions which is focussed on getting useful information on cascades. The interest in cascade damage has been growing along with the progress of development of fast breeder and fusion reactors, in which radiation damage due to high energy neutrons to a high fluence has been the major concern. Although the objective of in situ observation of heavy ion radiation damage is not necessarily restricted to the cascade problems as explained in a later section, we shall mainly confine ourselves in the present paper to deal with the cascade and its associated problems.

It has been well known that a primary knock-on atom (PKA) with energies well above the displacement energy will produce a series of displacement collisions within a limited volume. The resulting region with high density of displacements is called cascade. It was first treated both by theory and by computer simulation [1–5]. Early experimental evidences by electron microscopy and field ion microscopy techniques have shown that the cascade may be divided into subcascades [6–8] if the energy of the PKA is higher than a certain critical value [6,7]. The structure of the cascade has been considered inhomogeneous in such a way that the center is richer in vacancies with interstitials in the periphery [9,10]. It has been further demonstrated by a molecular dynamics computer simulation that the central vacancy rich core may be in a liquid-like state at the end of the collisional phase of the cascade which is initiated from a high energy PKA [11].

A number of new problems have evolved with the progress of knowledge of the structure of the cascades.

(1) What fraction of the total displacements will survive after the cooling of the cascade?

(2) What fractions of the survived point defects form clusters?

(3) What is the mechanism of energy dissipation during the cooling phase, particularly with regard to the energy deposited in electronic excitation during the collisional phase of the cascade?

(4) Are the cascade-originated biases as 'production bias' [12] or CLIB (cascade localization induced bias) [13,14] really as substantial as these theories predict and what are the quantitative consequences?

(5) Are interstitial loops formed directly from the cascade and what are the mobilities of interstitial clusters or loops?

All of these are essential and fundamental problems for the quantitative evaluation of radiation damage to be clarified in more detail by experiments. They are dependent on the PKA energy and on irradiation temperature. In situ observation of heavy ion radiation damage is expected to have a potential to give useful information for at least some of these important issues.

2. Historical development of in situ observation techniques and facilities

Historical development of in situ observation of heavy ion radiation damage will be summarized here. A list of facilities of in situ observation of ion radiation damage has been given in a previous publication [15].

Most of the facilities comprise an electron microscope and ion accelerator(s), but the first trial was made by using O^- ion beams generated at a $BaCO_3$- or $SrCO_3$-coated electron gun filament inside the electron microscope [16]. Connecting an ion accelerator to an electron microscope was first made by researchers at Harwell [17,18], and then this was followed by research groups at the University of Virginia, University of Tokyo, Argonne National Laboratory and so on. Some of the facilities utilize a high voltage electron microscope (HVEM), which adds additional capability to the facility. In the 1980s, various facilities have been built mainly in Japan, but their major objectives are slightly different from each other.

These objectives listed below include the new problems listed above.

(1) Damage microstructure evolution by energetic heavy ions in comparison with that by energetic electrons in a high voltage electron microscope.

(2) Mechanism of helium embrittlement or of radiation embrittlement in general with a special attachment to break the specimen inside the electron microscope.

(3) Mechanism of helium bubble formation.

(4) Studies of cascade damage by heavy ions, in particular, by self ions.

(5) Direct comparison of electron and ion radiation damage using an accelerator–HVEM link.

(6) Fusion radiation effect studies using dual ion or triple ion (dual ion and high energy electron) beams.

(7) Mechanism of radiation induced mixing and radiation induced/enhanced phase transformation.

(8) Radiation induced or enhanced diffusion.

(9) Ion implantation effects simulating the effect of nuclear transmutation.

(10) Ion induced amorphization.

(11) Studies of the effect of electronic- to nuclear-stopping power (ENSP) ratio [19] on microstructural evolution in semiconductors and insulating ceramics.

The accelerator–HVEM link has a special advantage allowing us, aside from direct comparison of electron and ion irradiations, to determine the nature of the ion irradiation induced defect clusters by irradiating electrons with energies above the displacement threshold [20].

Some of the problem areas listed above are very interesting though extensive studies have not been done so far.

Neither included in Ref. [15] nor in the objectives listed above are a range of research subjects in plasma–wall interactions as exemplified by mechanistic studies of blistering. Several dedicated facilities have been constructed for these objectives [21,22]. Some of the facilities listed in Ref. [15] have been used for these purposes as well.

3. Experimental facility for in situ observation at the University of Tokyo

The experimental facility we have been using comprises a 400 kV heavy ion accelerator and a 200 kV electron microscope, the details of which have been reported elsewhere [15,23,24]. Recently, the electron microscope has been renewed to a JEM 2000FX but the results given here were obtained using the original microscope, JEM 200C installed in 1976.

Major capabilities of the facility are summarized in Table 1. As shown in this table, the facility can provide energetic ions of practically all elements including metal ions up to 400 keV which bombard the specimen in the electron microscope in the temperature range from 120 to about 800 K during in situ observation. Even though we have used various kinds of specimens, in the present report discussion will be focussed on the results obtained in gold.

4. Defect accumulation: defect production and annihilation

In this section, we shall discuss the defect production and annihilation processes during heavy ion irradiation. Defect accumulation is an important process which, in the case of pure metals, can be conceptually reduced to the difference between defect production and annihilation. It is a great advantage of in situ observation that both production and annihilation processes of each individual observable defect can be determined dynamically during irradiation.

4.1. Evolution of microstructure to a high dose

Defect clusters formed during irradiation can be observed continuously using the facility. To obtain general microstructural evolution characteristics, microstructure at high dose rate (10^{15}–10^{17} ions/m^2 s: 10^{-4}–10^{-2} dpa/s with 300-400 keV heavy ions, mainly Al^+, Ar^+, Ni^+ and Xe^+ ions) up to a high fluence has been continuously monitored in situ for various metals [25] as nickel, aluminium, vanadium and stainless steels at room temperature. There are common features in microstructural evolution for these metals at room temperature; first, black dot images are immediately observed after the start of irradiation. Soon some of them are identified as loops, which then grow to a larger size, interacting with each other to form tangled dislocations. Further irradiation converts the tangle into a regular array of dislocations along specific crystallographic orientations [26,27]. The resultant microstructure is similar to that of polygonization, which is very stable for further irradiations.

From the behaviour of their growth, the loops observed to grow in size in the high dose region are believed to be of interstitial type in nearly all cases. However, in the case of aluminium, both interstitial and vacancy types are existing together in the black dots at the early stage of irradiation. The nature of the point defect clusters is dependent on various parameters as thickness, temperature and dose. This will be discussed in the sections to follow.

The behaviour described above seems to be common to a homologous temperature range where interstitials have high mobility whereas vacancies are not very mobile. However, whether this behaviour is analogous to that observed in bulk materials may depend again on various

Table 1
Major capabilities of the in situ observation facility at the University of Tokyo

Heavy ion accelerator	
Accelerating voltage	20–400 kV
Ion source	Danfysik 911 A
Ions generated	almost all ions including metal ions with the vapour pressure exceeding 0.1 Torr at 1700°C
Analyzing magnet	225 MeV amu
Electron microscope	JEM-200C[a]
Specimen temperature	120 K–800 K
Imaging	silicon vidicon/30 ms per frame
Interface	
Incident angle of ion beam to the electron beam	45°
Beam deflector	electrostatic (0–15 kV/cm)
Capacity	up to 400 keV (single-charged ion)

[a]Recently renewed to JEM-2000FX.

factors which will also be discussed later. If one starts with annealed materials, most of the materials start with recombination dominant regime, except a very early stage of irradiation where direct production of clusters from cascade is dominant as described in Section 4.4, changing into dislocation sink dominant regime for further irradiations. Some of the materials, for example gold thin foil, even start with a surface sink dominant regime and never get into a dislocation sink dominant regime. Since the early stage of microstructural evolution will strongly affect the later microstructural changes, the microstructure after high dose irradiation will severely deviate from bulk irradiation with, for instance, neutrons to a high dose, so that we have stopped examining high fluence effects in thin foil specimens using this facility.

4.2. Observation of subcascade images; the dependence of irradiation temperature, materials and incident ions

Irradiation of thin foil specimens of gold, aluminium and SUS316 stainless steel with heavy ions at 120 K produces defect images to be related to the formation of cascade [28]. Major observations are summarized in Table 2. Observability of cascade images depends clearly on materials. In gold, interstitials are extremely mobile and annihilate very effectively at the surface in the thin foil, leaving vacancies behind. In aluminium, length of replacement collision sequence may be shorter according to the light mass, enhancing the probability of intra-cascade recombination. In the temperature region where both vacancies and interstitials can migrate, i.e., above stage III temperature, ordinary interstitial-dominant microstructure evolution becomes prevailing and vacancy clusters can only be observed in a very thin part of the specimen or very close to dislocations [29]. In austenitic stainless steels, interstitial mobility is considered to be low with migration energy of 0.9 eV [30,31]. Therefore, substantial intra-cascade recombination may be expected. It should be mentioned that during irradiation, defect migration may be enhanced by subthreshold energy transfer as observed in aluminium [32,33] so that the material behaviour under irradiation may be different from that expected based on the defect properties without irradiation.

Dependence on the incident ion mass at a given energy is also evident. Displacements may be more localized for heavier incident ions. This will provide interesting information on the relation between the visibility of vacancy cluster and critical vacancy density in the cascade. Using the data on the defect cluster formation in pure metals by heavy ions of several tens of keV, which have been accumulated for the past 15 yr [34,35], Morishita et al. have discussed the criterion for the clusters to be observable, concluding that the local vacancy density should exceed a critical value which is determined for each material [34–36]. This point will be discussed in more detail in Section 5.1. Visibility and invisibility of defect clusters in aluminium for irradiations at 120 K with Xe^+ and Al^+ ions, respectively, can be explained by the concept of local vacancy density. As discussed in the previous paper [28], another possibility should also be considered: In the case of Xe^+ ions, having a shorter range than Al^+ ions, fast moving interstitials have a greater probability to be annihilated at the surface and more vacancies can accumulate to form vacancy clusters. Recent results on vanadium using V^+ self ions and Au^+ ions [37] may be explained along these lines. Satoh et al. [38] have discussed the criterion of subcascade formation, intersubcascade separation or on subcascade overlapping by considering mean free distance of primary collisions and the range of the recoil atoms. Similar consideration may be applied to the case of the dependence of defect cluster formation on the mass and energy of incident ions.

As discussed in the following section, the visibility of vacancy clusters is strongly dependent on the specimen

Table 2
In situ observation of heavy-ion radiation damage at 120 K in gold, aluminium and SUS316 stainless steel [28]

Material	Irradiation	Observation
Au	80, 177, 400 keV Xe^+; 181, 400 keV Al^+	● group of clusters with comparable numbers of incident ions
		● the number of subcascades increases with incident ion energy
		● Xe ions produce more subcascades than Al ions
		● no significant change by annealing to room temperature
		● irradiations at room temperature and at 473 K give similar cascade images
Al	400 keV Xe^+	● small dots with number density comparable to incident ions
		● no cluster images are left after annealing to room temperature
	400 keV Al^+	● no cluster images are observed
		● no cluster images are observed for both Xe and Al ion irradiations at room temperature[a]
Stainless steel	400 keV Xe^+	● dot images with number density much less than the number of incident ions
	400 keV Ar^+	● too coarse image to be identified as individual cascade

[a]After a certain incubation dose (~ 10^{19} ions/m^2), dot images appear. Most of these images are of interstitial type but a few vacancy type clusters appear near the edge of the perforation of the TEM specimen, where thickness is small [29].

thickness. It might be necessary to re-examine the defect yield values in the literature in terms of the specimen thickness.

4.3. The effect of specimen thickness

The importance of the role of surface sink is dependent on the target materials, irradiation temperature and incident ions. In fact in some cases, heavy ion irradiations produce similar microstructural evolution as in the bulk material, but in other cases, heavy ion irradiations give rise to quite different microstructural changes due to a strong sink effect of surface for mobile point defects. Spatial distribution of induced damage and of implanted incident ions will add additional complexities.

The thickness effect has been well demonstrated by using a wedge-shaped specimen in nickel [39,40] irradiated at 570-770 K with 300 keV Ar ions, in which thicker part (thicker than ~ 200 nm) shows the microstructural evolution similar to that observed in bulk specimens, whereas in a very thin part, vacancy clusters presumably formed by a single cascade event are observed. However, their visibility is very limited. There is a narrow region with intermediate thickness where point defect annihilation balances, not producing any point defect clusters at all. As the irradiation proceeds, these specific zones shift by growing dislocation loops and tangles in the adjacent regions, thus by changing the dislocation sink strength. Cascade-induced metastable vacancy clusters can be observed in a relatively dislocation-free volume. This thickness dependence of microstructural evolution in nickel is schematically shown in Fig. 1 [40].

From the observation given above, we may have a chance to observe cascade induced vacancy clusters by selecting appropriate specimen thickness and associated ion species and the energy, under the additional restriction that the cascade size is within the specimen thickness. Cascade efficiencies published so far [34,35] are obtained under such restrictive condition.

These considerations lead to the conclusion that gold is one of the extreme cases, in that surface sink dominant thin foil condition holds for a rather wide range of the specimen thicknesses. Kiritani et al. [41] also observed that in thin foils of copper, silver and Cu_3Au irradiated with 120 keV self ions the majority of defect clusters are of vacancy type. In other words, cascade characteristics can be derived through the observation of vacancy clusters in thin foil specimens.

4.4. Observation of single cascade events

The term 'single event' is sometimes used as a phenomenon taking place during and after a very short pulse of radiation. This is a different terminology from what we are intending to use here, because we are observing the cascade event produced by a single incoming ion. This is a

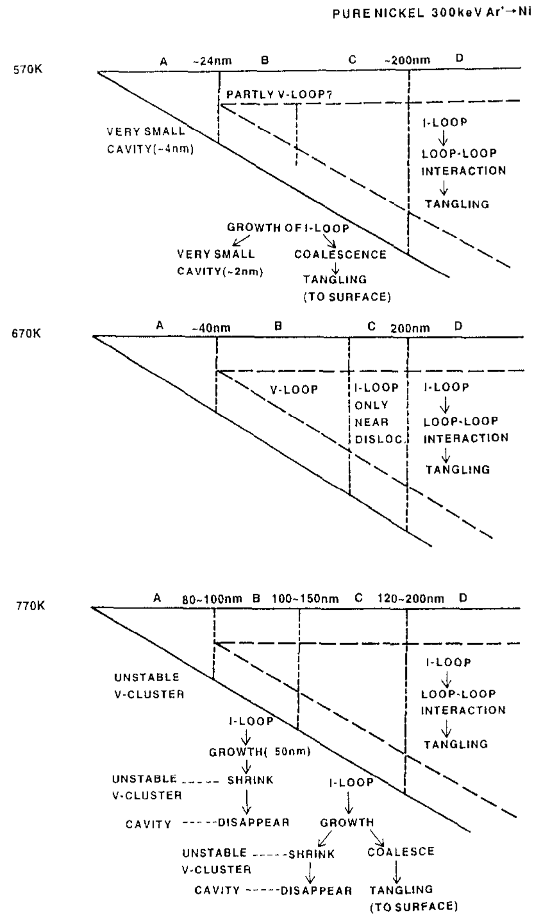

Fig. 1. Specimen thickness dependence of microstructural evolution in wedge-shaped nickel specimen during irradiation with 300 keV Ar^+ ions at 570, 670 and 770 K. There are at least four characteristic regions, A, B, C, D, where evolution in region D is similar to that in the bulk specimens [40].

very unique feature of in situ observation of heavy ion radiation damage. We have been using mostly thin foil gold specimens intentionally for the purpose of observing single cascade events and cascade–cascade interactions.

Observation of accumulation of vacancy clusters in gold at 300, 473 and 573 K during irradiation with 400 keV Xe^+ ions, which will simulate gold self ions, shows that the cluster density is proportional to the ion fluence up to 10^{14} ions/m^2 [15]. However, annihilation of clusters begins to occur at a fluence of 10^{14} ions/m^2 and above. Therefore, isolated cascades can be observed only with a very low irradiation fluence. It is a great advantage of in situ observation that production rate and annihilation rate can be determined separately. In this experiment, we have observed the defect accumulation continuously by videorecording, without stopping the beam for intermediate analyses, to avoid the possible effect of irradiation interruption found in austenitic stainless steel [26]. This sacrifices the precise determination of the cluster yield, because the diffraction condition is kept constant. Here, the cluster yield, sometimes called the defect yield, is defined as the number of defect clusters formed per incident heavy ion. Several defect clusters are frequently observed to be formed simultaneously in a group. The number of groups of defect clusters formed per incident heavy ion is called the cluster–group yield, or simply the group yield. The cluster–group yield of 0.4 was low as discussed in Ref. [15]. There was a suspect that some of the defect clusters were not observed under a constant diffraction condition in continuous in situ observations. Since irradiation interruption is found to have no effect in the present case, precise defect yields, i.e., cluster–group yield and cluster yield have been determined using self ions with energy range of 20–400 keV for low fluence [42] in which isolated cascade conditions are fulfilled. Self ions are assumed to simulate the PKA of the same energy. In these experiments, ion beams were switched off during TEM observations and the defect images were observed using several diffraction vectors. The group yield is found almost unity for the energy range of incident ions of 100–400 keV, whereas the cluster yield approaches nearly 2 at the energy of 400 keV. Probability function of the number of clusters in a group extends to 7 clusters in a group as shown in Fig. 2(a). Relatively large scatter of the data is due to the difficulty of getting enough statistics in order to assure to collect single cascade events. The data were taken only within a very low fluence region of the order of 10^{14} ions/m^2 so that the number of clusters was small. Using these probability functions together with calculated PKA energy spectrum for 14 MeV neutrons (Fig. 2(b)), we can calculate out the distribution of the number of vacancy clusters in a group [42], which is in reasonable agreement with Kiritani's results [43] obtained in thin foil gold irradiated with 14 MeV neutrons from RTNS-II (rotating target neutron source-II) as shown in Fig. 2(c). Similarly, the low fluence data obtained in thin foil gold irradiated in a fast neutron source reactor, YAYOI, can be explained using the same probability functions. The analyses have further been extended to high energy self ion irradiations [44]. A review of the recent results is given in a review article by Sekimura [45].

4.5. Cascade–cascade interaction

Linear relation between the cluster density and ion fluence holds only at the early stage of irradiation. The cluster density tends to deviate from linearity to half power of the fluence and after some transient stage it approaches to a saturation value [15]. This process is discussed from the defect kinetics and is modelled to be separated into four stages by Kiritani [46]. In this model, there is a stage with square dependence of dose, which is ascribed to be due to the formation of vacancy clusters with the help of other cascades [43].

In 14 MeV neutron irradiated gold, there is an intermediate fluence region where the exponent of the cluster density growth curve is higher than one between two linear regions [43]. Similar region is also found in the specimens irradiated in the YAYOI reactor as well as those irradiated with 20 MeV self ions [45]. This phenomenon is characterized by the formation of new small sized clusters near the pre-existing group of clusters and is considered to be due to the effect of nearby cascade.

It has been often observed that defect clusters tend to be generated near the pre-existing clusters as exemplified in fig. 1 of Ref. [28]. It is reasonable to assume the existence of invisible latent clusters which will become

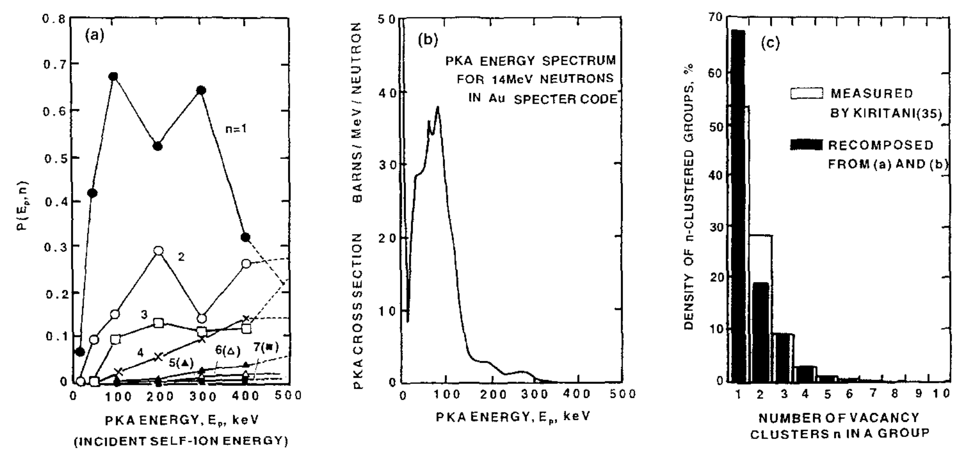

Fig. 2. (a) Experimentally determined probability function of the number of vacancy clusters in a group in gold as a function of incident self ion energy which is regarded to PKA energy, for a low fluence region where no cascade overlap occurs. (b) Calculated PKA energy spectrum in gold for 14 MeV neutrons by the SPECTER code. (c) Comparison of the distribution of the number of vacancy clusters in a group experimentally obtained in gold thin foil irradiated with 14 MeV neutrons with RTNS-II by Kiritani et al. [43] and that calculated by the probability function (a) and the PKA spectrum (b) [42].

visible by agitation from nearby cascade. The nature of this agitation has not been clarified yet. Energy transport by shock waves, ballistic energy transfer, energy transport by replacement collision sequences and focussing collisions are among the possibilities to be considered. By analyzing the experimental results, Sekimura has derived the minimum interaction energy of a PKA to produce new visible defect clusters to be 165 keV [45].

4.6. Stochasticity of defect formation and annihilation processes

In the case of heavy ion irradiation, heterogeneous defect cluster formation is often observed, particularly near pre-existing dislocations. It has been often observed in the case of heavy particle irradiations including neutron irradiations that dislocations become decorated with small loops, which are identified as interstitial type in nature. Muroga et al. have examined shrinkage of quench-induced vacancy loops in an Al–0.006% Ge alloy by 400 keV Al^+ ion irradiation, finding that the vacancy loop annihilation takes place rather discontinuously [29]. Immediately after Al^+ ion irradiation, the pre-existing faulted loops introduced by quenching turn out to be perfect loops. Then the loop edge is decorated by small loops and the original loops eventually annihilate catastrophically. Such a remarkable phenomenon has never been observed in the loop annihilation process by electron irradiation and is believed to be characteristic to cascade damage. Nature of the small loops has not been identified. They might be of interstitial type, originated from cascade capable of making long range migration, being trapped by the strain field of pre-existing dislocations or loops.

As mentioned in a previous section, annihilation process of vacancy clusters induced by heavy ion irradiation in gold begins to occur at a relatively low fluence of the order of 10^{14} ions/m^2. When the annihilation rate becomes equal to the production rate, steady state concentration will result. This condition is reached at a fluence slightly exceeding 10^{16} ions/m^2. The process of annihilation is stochastic in nature [47,48]. The clusters, once formed, seldom grow to a larger size, except in a very special case, in which two or more cascades overlap with each other and the cluster size abruptly increases [49]. At irradiation temperatures of and above 473 K, the vacancy clusters take the clear form of either loops or stacking fault tetrahedra, the former being less stable [50], and after keeping the specimen for long time without irradiation, most of the clusters left are stacking fault tetrahedra. The cluster annihilation process is mainly due to the absorption of interstitials coming from nearby cascades, because the lifetime of the clusters, which is a measure of their stability, is enhanced by two orders of magnitude when the ion beam is off [50].

The individual loop diminution rate is shown to be proportional to the local interstitial concentration. Sudden size diminution in a stochastic manner implies that interstitial arrival at the position of the vacancy cluster is a stochastic process. In other words, the vacancy cluster is serving as a probe to detect spacewise and timewise fluctuation of interstitial concentration packets coming from nearby cascades [48,51].

5. Discussion

5.1. On the visibility of vacancy clusters formed directly from cascade

Formation of vacancy clusters by so called 'cascade collapse' has been studied for many years and ample data have been accumulated in pure metals as to the defect yield for heavy ions of several tens of keV [34,35]. Morishita et al. have discussed the criterion for the clusters to be observable in a semi-empirical way. They first calculated cascades using the binary collision code MARLOWE for a variety of combinations of incident ions and target metals, where the defect yield data are existing. Then for each vacancy position, vacancy concentration within a spherical volume of radius $5a_0$ is calculated and the vacancy density distribution functions are calculated out as shown in Fig. 3(a). If one assumes that the vacancy clusters are more easily formed for higher vacancy density, critical vacancy density for the formation of vacancy clusters can be determined from experimentally determined defect yield values as shown in Fig. 3(b). Interestingly enough, the critical vacancy density determined in such a way becomes almost constant for a target metal regardless of the incident ion species and their energies [34,35]. In fact in Fig. 3(b), the critical vacancy density is nearly constant to be 2.0 ± 0.3, except for the cases of 20 keV and 50 keV, where some of the defect clusters are too close to the surface. The overall result does not depend sensitively on the choice of the radius of the spherical volume over which the vacancy density distribution functions are calculated.

It is interesting to consider the reason why the critical vacancy density changes with target materials. As reported in Refs. [35,36], the critical vacancy density is in good correlation with the vacancy mobility at the melting temperature. This seems to suggest that the total number of jumps during the cooling phase of the cascade may have close correlation with the vacancy clustering. In fact, higher number of jumps result in lower critical vacancy density. Morishita [36] has also calculated the temperatures of the electron and lattice systems by solving a set of thermal transport equations taking into account an appropriate electron–phonon coupling constant, obtaining results for the initial kinetic temperature of 3500 K that for systems with high defect yield as Cu, Ag and Au, the lattice temperature remain to be above the melting point

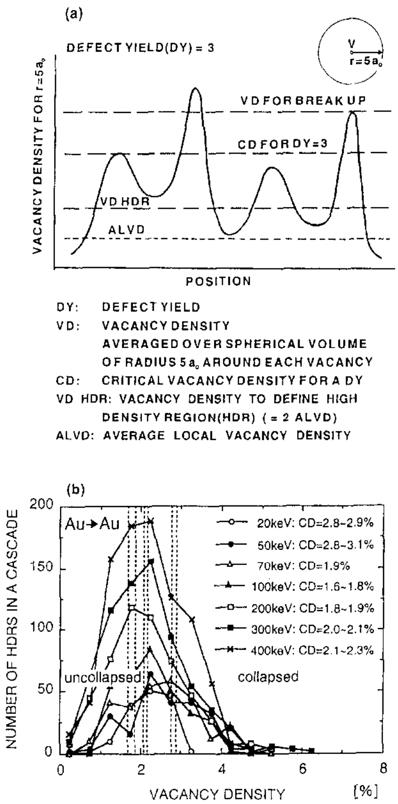

Fig. 3. (a) Schematic representation of vacancy density distribution within a cascade. Critical vacancy density corresponding to experimentally obtained defect yield is shown. (b) Vacancy density distribution function in terms of the number of high density regions (HDRS) in a cascade at the end of the collisional phase obtained by averaging 100 cascades from BCA calculations in self ion irradiated gold [36]. Assuming higher density will result in 'vacancy collapse', experimentally obtained defect yield will give a critical vacancy density for vacancy cluster formation [34–36].

for a few tens of picosecond while those with low yield as Ni, Fe, Mo and W, the lattice temperature goes below the melting temperature within about a picosecond or less.

In the case of iron, no vacancy clusters are observed, suggesting that the corresponding critical vacancy density should be very high to obtain such clusters. However, for higher incident ion mass, the vacancy density distribution curve seems to shift to higher vacancy density. This suggests that vacancy clusters might be observed if one uses heavier ions. In this regard, recent results on vanadium using V^+ self ions and Au^+ ions [37] are interesting because of the similarity of iron and vanadium: They detected very few clusters in the specimens irradiated with 200 and 400 keV V^+ self ions, while vacancy clusters were observed in a very thin region of the specimens irradiated with 50-400 keV Au^+ ions. This may be explained by the concept of critical vacancy density.

5.2. Geometrical effects: range of incident ions and specimen thickness

As mentioned earlier, the energy region of interest for the PKAs in fission and fusion reactor irradiations is roughly below 1 MeV. For self ions of this energy range, the range of particles is less than several fractions of a micron. For in situ TEM observations, the specimen thickness is in the same range, even if one uses a HVEM for the observation.

Thickness of the specimen modifies the relative importance of the surface sink effect of the specimen. The role of surface sink is dependent on the target material, irradiation temperature and incident ion. The variety of the phenomena due to thickness may be grouped into several categories by scaling it in terms of a certain parameter related to the separation of major sinks, as for example, in terms of the specimen thickness divided by a certain point-defect related length parameter, which we tentatively assume to be the mean free length of interstitials before being annihilated. This should be related to the local sink strength for point defects. The thickness domains may be grouped into four groups as shown in Fig. 1. The kind of observable defects changes according to the specimen thickness, i.e., surface sink and to the state of other point defect sinks. The situation is schematically shown in Fig. 4. The thickness of the specimen together with the state of other sinks should be carefully evaluated to draw any conclusion. For the study of cascade structure through vacancy clusters, surface sink effect for interstitials must be dominating. The condition may be described such that the mean free path for the interstitials before annihilation λ should be comparable to the specimen thickness, t, thus

$$\lambda \sim \lambda_{is} \sim t, \qquad (1)$$

where λ_{is} is a mean free length for interstitials to annihilate at the surface. The probability of annihilation of interstitials at vacancy clusters may be low at the initial stage of irradiation at relatively low temperature because both the concentration of vacancy clusters is low and the interstitial motion may be more or less one-dimensional. Trinkaus et al. [52] have given the reciprocal mean free path of one-dimensionally moving interstitials to be

$$\lambda^{-1} = \Sigma \sigma_j C_j + \pi \rho d/4, \qquad (2)$$

where C_j is a number density of defect j as an interstitial sink, σ_j the effective interaction cross-section, ρ a dislocation density and d the effective interaction diameter. Neglecting dislocations, the first term of the right hand side of Eq. (2) is dominated by the surface.

On the other hand, during irradiation at elevated temperatures, the mode of vacancy cluster annihilation is mainly by absorbing interstitials [47,48]. This is because the total cross-section of vacancy clusters for interstitials becomes larger and possibly because fraction of three-dimensional motion of interstitials may become larger. A

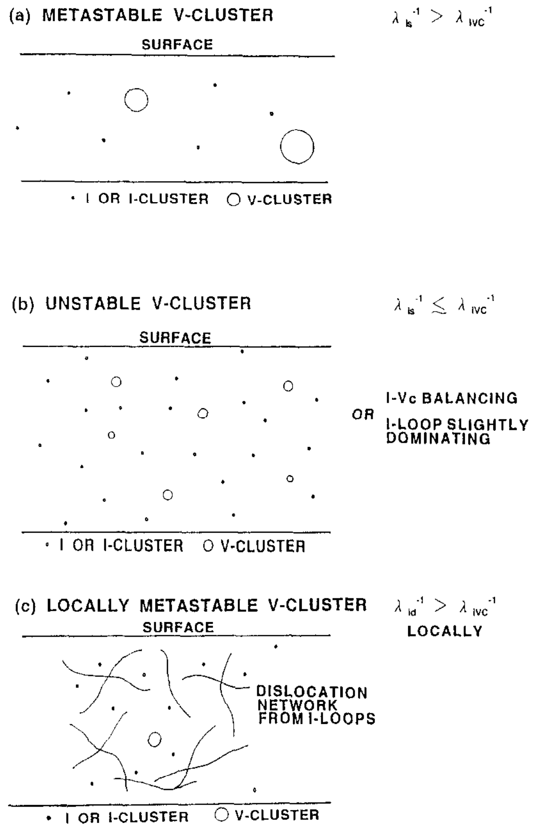

Fig. 4. Schematic representation of (a) the 'thin foil condition' to observe vacancy clusters created in a cascade. (b) Condition of no observable defects and (c) that of formation of metastable vacancy clusters at some stage of extended dose are also shown.

condition will be reached, where balance between vacancy clusters and interstitials is established and newly formed vacancy clusters are destroyed immediately by the flux of interstitials as shown in Fig. 4(b). At very low temperature where interstitial mobility is reduced, interstitial supersaturation may increase, resulting in the formation of interstitial loops even in a thin gold foil [53].

In nickel irradiated with 300 keV Ar ions at 770 K, metastable vacancy clusters with lifetime of 1-30 s are observed as described above (Section 4.3) [24,39,40]. In the region where the metastable vacancy clusters are observed, the effective radius of the region is probably similar to the above mentioned mean free length of interstitial annihilation at the dislocation wall surrounding the region. This situation is schematically shown in Fig. 4(c).

The discussions given above may indicate that some of the cascade characteristics can be derived through the observation of vacancy clusters, provided appropriate conditions as to the specimen thickness and associated ion species and the energy are selected with an additional restrictive condition that the cascade size should be within the specimen thickness.

The condition that the cascade production is confined within the TEM observable thickness should be satisfied for an incident ion to be representative of a PKA of the same energy. The energy of 400 keV of self ions in gold for an electron microscope with accelerating voltage of 200 kV is almost the upper limit in this context for normal incidence. In fact, complex situation comes in in the case of 20 MeV and 200 MeV self ion irradiations [44], even if thin foils are used by stacking several foils to match the range of impinging ions. The complexity arises from ejection or from implantation of secondary recoils to and from the adjacent foils. Such effects must be taken into account in the thin foil experiment using neutron or high energy ion irradiations as well. Kiritani et al. have used inclined incidence of impinging ions for the study of accumulation of vacancy clusters [41]. This technique allows us to change relative importance of surface sinks without changing the energy of the impinging ions.

For low energy self ions, the overlap of the cascade volume with the free surfaces results in the reduction of the defect yield. Corrections have been made to adjust this effect [42] to obtain the probability functions of the number of clusters in a group as a function of PKA energies as described in Section 4.4.

5.3. Nature of the observed defects

In the self ion irradiation of gold up to the incident energy of 400 keV, the defects observed are found to be of vacancy type by TEM image analysis and their high temperature annealing characteristics [43]. Dark field images with (200) reflections were taken for the analyses. The nature of the clusters was determined using an inside–outside contrast technique. On the other hand, in dynamic in situ observations, mostly carried out using bright field kinematical condition, it is difficult to determine the nature of the defects during ion irradiation. In gold, it is almost certain that the defect clusters observed are of vacancy type, judging from the kinetics of their annihilation. However, in copper or in nickel, for example, both vacancy and interstitial types of clusters are existing depending on the thickness of the specimens. In general, it seems necessary for the discussion of the nature of the clusters to examine the dependence of specimen thickness, temperature and fluence. Application of a HVEM irradiation for a very short duration to assist the determination of the nature of the ion-induced defect clusters [20] is useful when the HVEM is available.

5.4. Currently confronting experimental limitations

5.4.1. Experiments for higher PKA energies

In the case of gold, surface sink dominant regime seems to hold for the specimen thickness of roughly 200 nm. If this regime extends to thicker specimens, cascade structures with the PKA energies above 400 keV may be examined for normal incidence of impinging ions. In this regard, it is interesting to examine whether the results obtained for incident self ions with energies up to 400 keV can be reconstructed from the calculated secondary knock-on energy spectrum combined with the probabilistic functions described in Section 4.4. Such kind of methodology may be required to investigate the effect of cascades with the PKA energies up to about 1 MeV, which are expected in radiation effects with D–T fusion neutrons. It is also very interesting to examine the effect of oblique incidence of impinging ions.

5.4.2. The effect of cascades in bulk materials

The application of in situ observation techniques to the study of radiation effects in bulk materials is one of the greatest concern to us. In some of the materials as nickel and stainless steels, cascade-induced vacancy clusters cannot be observed in the in situ observation experiments using a foil of ordinary thickness, say 200 nm, at least at the early stage of irradiation in the temperature range where interstitials are mobile. Instead, after a short incubation time, black dot images are formed and then interstitial loops are growing as observed in ordinary bulk specimen irradiations. In situ observations in these materials should be compared with those of bulk materials irradiated with self ions of higher energies and with fast neutrons, though there are some experimental issues as the difference of displacement rates, degradation of materials by surface oxidation for prolonged irradiations at high temperatures and so on, that will make direct comparison difficult.

5.4.3. Nascent cascade state at low temperature

Currently, the lowest temperature available in our facility is about 120 K. As discussed in Section 4.2, there are definitely temperature dependence of defect cluster formation on irradiation temperature. It is very interesting to perform in situ self ion irradiation experiment at very low temperature where point defect thermal mobility is very low. It is worthwhile to relate such observations with those of 14 MeV neutron irradiations obtained by Shimomura et al. using a cryotransfer technique [53–56] to a TEM without raising the temperature. The most intriguing subject is whether interstitial clusters are formed directly from the cascade, thus surviving high temperatures. This should have a strong impact on later microstructure evolution as mentioned in Section 1. In carrying out cryogenic experiments, it will probably be necessary to consider the temperature dependence of the effect of specimen thickness on the defect cluster stability as well.

6. Future prospect

Although a number of experimental difficulties are foreseen to be existing in the in situ observation techniques for the study of the nature and the effect of cascade, it is almost the only possibility to derive information of a single cascade event in crystals. Moreover, the technique will allow us to examine inter-subcascade or intra-cascade interactions, intercascade interactions and defect accumulation mechanisms not only in surface sink dominant cases but also in other cases if proper experimental conditions are set up as to target materials, temperature and geometrical factors as thickness, incident angle and so on. Study of the dependence of crystallographic orientations combined with oblique incidence technique may give useful information on the motion of the point defects and their clusters.

Further development of experimental techniques will be required, e.g., extending irradiation temperature range from cryogenic temperature to high temperature region where defect clusters become unstable, extending ion energy range preferably to a MeV region with accompanying technical development as introducing HVEM and establishing inclined incidence of ion beams, improving ion beam measuring system to extend the experiment to a very low ion current region, increasing versatility of the accelerating voltage of the electron microscope allowing displacement damage to occur to facilitate determination of the nature of defect clusters, improving vacuum to allow prolonged irradiation without degrading the specimen and so on.

The recent trend of in situ observation technique is to install various analytical tools to the electron microscope. This will enhance the capability of the in situ experimental technique. Developing 'a micro-radiation laboratory' as schematically shown in Fig. 5 is one of the important future directions.

Another point I would like to make is the importance of computational studies and modelling activities. In our experimental facility, we are able to observe the dynamic

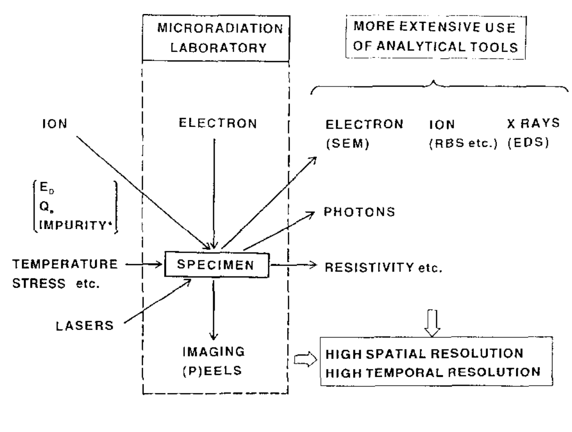

Fig. 5. The concept of 'micro-radiation laboratory' as a future direction of in situ observation facility. Various disturbances are applied to the specimen from which various responses are emitted. Here, ions will introduce nuclear energy deposition (E_D), electronic energy deposition (Q_e) and impurity atoms.

phenomena taking place within 30 ms. On the other hand, the cascade will come to quasi-equilibrium typically within 10 ps. Probably, the time resolution of the in situ observation technique may be improved by an order of magnitude, but still we have a wide gap over six orders of magnitude of time scale between molecular dynamics calculation and the in situ experimental technique. Recent advance in detecting extremely fast phenomena in femtosecond region [57] is giving us hope to observe cascade phenomena particularly through optical measurements but this will require revolutionally ingenious idea. For the moment, for microstructural evolution, lattice Monte Carlo calculations [58,59] taking the results of binary collision approximation (BCA) or molecular dynamics (MD) calculations as an input [60] will be useful to bridge the gap between the nascent cascade calculations and microstructural observations.

Finally, I would point out that in situ observation of ion radiation damage will be used as a verification tool of computational studies. Recent status of the verification of large scale MD calculations on cascades by experiments still remains to be distant from full satisfaction, leaving a number of uncertainties in the calculated results. In situ observation of cascade using thin foil is only for a special case which may be different from the cascade in bulk specimen. However, it will provide useful information to be used for the verification of MD calculations of cascade in a medium containing surface.

7. Conclusion

(1) In situ observation of radiation damage with energetic heavy ions is a useful method of investigating radiation effects induced by high energy heavy particles as fast neutrons. For the past twenty years, various facilities have been constructed in the world and since the 1980s, a large number of facilities, each with slightly different objectives have been constructed mainly in Japan. The present status was reviewed.

(2) One of the major objectives of the in situ experiment is to elucidate the cascade damage produced by an energetic primary knock-on atom. Major results on the fundamental aspects of cascade damage have been reviewed. The facility we have been using allows us to observe a single cascade event. Such information cannot be obtained by other means.

(3) By collecting the data on vacancy clusters in the single cascade events for various incident self ion energies in gold, probability functions of the number of subcascades in a group are derived. Using these functions, distribution functions of the number of subcascades in a group are derived for the cases of irradiations with 14 MeV neutrons and with fission reactor neutrons at low fluence, knowing the PKA energy spectrum obtained by neutronics calculations. They are in good agreement with experimentally obtained distributions.

(4) For higher fluence, the accumulation of vacancy clusters deviates from linearity with the fluence, indicating the presence of intercascade interactions.

(5) Visibility of vacancy clusters reflecting the cascade characteristics depends on various factors. Particularly important are the effects of specimen thickness, state of point defect sinks and of irradiation temperature.

(6) Issues of the experimental technique are summarized and future prospects of this technique are discussed.

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