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Implications of defect clusters formed in cascades on free defect generation and microstructural development*

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Abstract.

A large fraction of the defects produced by irradiation with energetic neutrons or heavy ions originates in cascades, i.e., defects are spatially and temporally closely correlated during their formation. As a consequence, not only increased recombination of vacancy and interstitial defects but also significant clustering of like defects occur. Both processes reduce the number of point defects available for long range migration compared to that available when defects are produced as isolated pairs or in small groups. Some consequences of defect clustering in cascades will be discussed in a semi-quantitative form with the aid of calculations using a very simplified model: Quasi-steady-state distributions of immobile vacancy and/or interstitial clusters develop which, in turn, can become significant sinks for mobile defects, and, therefore reduce their life-time. Although cluster sinks will cause segregation and, potentially, precipitation of second phases due to local changes of composition, the finite life-time of clusters will not lead to lasting, local compositional changes. A transition from highly dense interstitial and vacancy cluster distributions to the void swelling regime occurs when the thermal evaporation of vacancies from small vacancy clusters becomes significant at higher temperatures. Unequal clustering of vacancies and interstitials leads to an imbalance of their fluxes of in the matrix and, hence, to unequal contributions to atom transport by interstitials and by vacancies even in the quasi-steady state approximation.

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1. Introduction.

A number of properties of materials, such as strength, plasticity, and thermal and electric conductivity, are sensitive to various aspects or components of the microstructure. Here we will use the term microstructure in a quite general sense: including the atomic arrangement, i. e., crystal structure (or the lack of it in amorphous solids), state of order, point defects, point defect clusters, dislocations, grain boundaries, spatial distributions of alloying elements, as well as precipitates of second phases and their relations to other microstructural features. The initial microstructure is established during the fabrication process which may include mechanical deformation, recrystallization, phase transformations and precipitation of second phases.

Exposure to irradiation by energetic particles modifies the initial microstructure not only by acceleration of mechanisms in action during ordinary use of the material, but also by mechanisms specific to irradiation, e. g., displacement mixing and disordering, and non equilibrium segregation of alloying elements resulting from preferential coupling of fluxes of alloying components to persistent point defect fluxes. The evolution of microstructure, already very complex during ordinary processing and use of materials, becomes even more complex in an irradiation environment because of opposing mechanisms and forces: for example, atoms are transported in the opposite and the same direction, respectively, by fluxes of vacancies and of interstitials; the usually small difference in fluxes of opposite types of defects results in void swelling, and in dislocation climb and creep; the difference in coupling of an alloying element to fluxes of vacancies and of interstitials leads to radiation induced segregation even when both defect fluxes have the same magnitude; radiation induced segregation is opposed by radiation enhanced diffusion which tends to bring the material closer to its thermodynamic equilibrium state.

The complexity of and the interaction between different mechanisms is the major reason that our understanding of the evolution of microstructures under irradiation has remained predominantly qualitative despite extensive research in this area over the past several decades. All the basic processes that modify the microstructure have been recognized for some time (see, e. g., reference 1). However, good quantitative descriptions of several of the individual processes are not yet available. Here, we will outline some of the present knowledge on "freely migrating defects (or free defects)", i. e., mobile defects, and their relation to the evolution of the microstructure of alloys.

Some key features pointing to the deficiencies in our quantitative understanding of microstructural evolution are found in the experimentally well established dependence of radiation-induced segregation on the mass of the ion used for the irradiation in the MeV energy range. For example, the efficiency of inducing segregation in dilute Ni(Si) alloys decreases dramatically from that of protons via that of He, Li and Ne ions to that of Ni and Kr ions. [2,3] The efficiency of the last two ions is only a few percent of that observed for protons when compared at the same calculated dose in displacements per atom (dpa). Similarly, measurements of self-diffusion in Ni and diffusion of Ni in Cu show that only about 1.5% of the calculated displaced atoms contribute to radiation-enhanced diffusion during 300 keV self-ion irradiation. [3, 4] The low efficiencies for heavy ions have been related to the fact that a large fraction of the defects are produced in close proximity to each other in displacement cascades.

In the following section the defect production process will be described with special emphasis on the spatial and temporal correlation of defects produced in cascades, the recombination of vacancies and interstitials, and clustering of like defects. Section 3 will be devoted to discussion of methods to obtain effective medium values of free defect fluxes in the presence of a developing defect cluster population. The free defect fluxes can, in turn, be used to describe the evolution of other selected microstructural features. Section 4 will illustrate the development of interstitial and vacancy clusters at relatively low temperature (below peak swelling) under simplified defect production conditions, i. e., all defects are produced in cascades of uniform size, and under the assumption that defect clusters above a minimum size are immobile. In the last section we will outline areas in which significant progress needs to be made in order to provide the basis for a reasonably quantitative description of microstructural evolution during irradiation.

2. Defect production.

Energetic particles passing through a solid lose energy by three distinct mechanisms: (i) elastic collisions with nuclei in which a kinetic energy, T, is transferred to lattice atoms, (ii) interactions with electrons which are mostly inelastic in nature, i. e., electronic excitations, and (iii) nuclear reactions. For recent reviews see references [6, 7]. All three processes mentioned can lead to defect production in the solid. The characteristics and importance for defect production depend on the type, mass and energy of the incoming particles, and on the characteristics of the solid. Elastic collisions lead to defect production in all solids provided the transferred energy exceeds the minimum threshold displacement energy. Electronic energy losses in metallic systems are largely dissipated in the form of heat without producing lattice defects except in case of charged particles with very high, GeV range, energies. However, electronic exitations can be important for defect production in ionic or insulating materials. Nuclear reactions may produce transmutation products, which can be considered as defects, but also produce defects by displacements via the recoiling nucleus and energetic reaction products. Although we will limit our discussion to defect production by elastic collisions, which are the most numerous defects generated under irradiation with fast neutrons or ions considered here, we emphasize that defects formed via the other two energy loss mechanisms can become important because those defects are often produced predominantly in isolation or in small clusters and, hence, contribute very efficiently to the freely migrating defect population as discussed by Rehn and Birtcher [8].

The spatial distributions of defects are best discussed in terms of the primary recoil spectrum produced by the incoming particles. When an atom in a solid receives, from the incoming particle, a transferred kinetic energy T larger than the threshold displacement energy, E_d, it will leave its original site. Such an atom is commonly referred to as a primary knock-on atom. Its energy is dissipated by inelastic losses to the electrons of the system and by elastic collisions with other atoms of the solid which, in turn, may become 'secondary' knock-on atoms and produce higher generations of knock-on atoms. Nuclear reactions are not induced by knock-on atoms with the possible exception of light atoms with rather high knock-on energy.

Primary knock-on atoms (PKAs) of energy $T >> 2\ E_d$ produce cascades of displaced atoms that are characteristic of the material and the PKA energy, but independent of the manner in which the PKAs were produced. PKAs of a given energy generate an average number of defect pairs, N_o, which can be calculated by well established binary collision codes. [9] In fact, it has become customary to quote the irradiation dose by the total number of displacements per atom (dpa) calculated by this NRT procedure.

The characteristics of PKA spectra resulting from different irradiation environments have been discussed on many occasions, see, e.g., Rehn and Birtcher [8] Averback et al. [10], or Wiedersich [11]. MeV type electrons produce only low energy PKAs which in turn generate mostly isolated defect pairs. Most of the PKAs for MeV light ion irradiations generate defect groups with less then 10 defect pairs. Heavy ions and moderated fission and fusion neutrons produce cascades of defects distributed over a wide size range, with the average size increasing with the hardness of the spectrum. 80% of the PKAs resulting from 14 MeV neutrons produce cascades with more than 100 defect pairs.

There are two basic processes which reduce the number of defects initially created in cascades to that which undergo long range migration, i.e., contribute to microstructural changes requiring diffusion. (a) recombination of vacancies and interstitials during the collisional and cooling phases of the cascade; and (b) like defects agglomerate within the cascade region to form immobile defect clusters in the form of small dislocation loops, stacking fault tetrahedra and other three-dimensional defect clusters. In addition, the defect clusters created act as sinks for migrating defects and, therefore, reduce the average number of defect jumps between creation and annihilation which, in turn, reduces their contribution to diffusional processes.

The process of formation and the arrangement of the defects after the cascade regions has returned to ambient temperature as derived from experimental observation and extensive model calculations has been reviewed recently (see, e.g., English et al. [12] and Diaz de la Rubia and Guinan [13]). Here we will summarize only some of the salient features.

The energetic primary knock-on atom distributes the damage energy (PKA energy minus the cumulative inelastic losses) through a quick succession of generations of knock-on atoms and replacement collisions in a few tenth of a picosecond within a small region of the crystal. The highly disordered state, and the density of kinetic energy (exceeding significantly that of the molten material at the melting temperature) in the central region of the cascade, suggests that this region at the end of the collisional phase, is best described as a highly superheated melt under compression. During the 'cooling phase' lasting from a few tenth of a ps to several ps, the excess kinetic energy is dissipated into the surrounding matrix. The undercooled 'melt' crystallizes in metals, presumably from the periphery towards the center of the zone. Defects inside the molten zone lose their identity during the melt phase. However, a net number of vacant lattice sites will remain after crystallization, corresponding to the net number of atoms ejected into the surrounding crystalline matrix during the cascade formation process. During the cooling phase, defect configurations rearrange. This includes recombination between opposite types of defects as well as clustering of like defects. Vacancy clusters form predominately in the central regions, whereas interstitial clusters form mainly in the peripheral regions of cascades.

The description thus far is based exclusively on model calculations, predominately on molecular dynamics calculations, since experimental information on the cascade formation has not been obtained in real time because of the extremely short time scale involved. Diffuse x-ray scattering at room temperature after low temperature neutron irradiation shows the presence of vacancy clusters and of interstitial clusters in Ni and Ni-alloys. [14] The vacancy clusters must have formed during the cascade events, because the thermal motion of vacancies during warm-up to room temperature is negligible. However, the interstitial clusters could have formed during warm-up. Rauch et al. and Peisl et al. have shown that small interstitial clusters exist below stage II annealing after fast neutron irradiation at liquid He temperatures in Cu and Fe, but not in Al or dilute Al alloys. [15-17]

Extensive evidence from transmission electron microscopy (TEM) exists that vacancy clusters in form of dislocation loops and/or stacking fault tetrahedra are present at low to intermediate specimen temperatures after cascade damage in many metals and alloys (see, e. g., English and Jenkins [18] and Kiritani [19]). Therefore, clustering of vacancies and collapse to loops or tetrahedra occur during the cascade event. It is highly likely that void-like clusters are also formed since the 'yield' of defect clusters visible by their strain fields is often considerably below one per cascade. Void-like clusters may collapse to visible loops during a subsequent, nearby cascade events as indicated by a more than proportional increase of visible defects with dose at low doses. [20] At higher doses, the defect yield becomes less than proportional to fluence as the consequence of cluster dissolution by overlapping subsequent cascades. [21] Gradual shrinkage and dissolution of vacancy clusters, presumably by absorption of an excess of mobile interstitials, has also been observed. [22]

Interstitial loops formed in cascades at low temperature have not been seen by TEM. This probably is a consequence of the small size of any interstitial cluster formed directly during the cascade event. The frequent observation of well resolved interstitial loops after irradiation with neutrons under conditions where interstitial concentrations in the matrix are too low for homogeneous nucleation of loops can be taken as indirect evidence for interstitial cluster formation during cascade events. [19,20]

When the energy of PKAs exceeds 15 to 20 keV, cascades tend to split into subcascades as evidenced by groups of visible defect clusters in close proximity to each other. [20,23]

3. Models for Free Defect Production.

A number of suggestions have been made to explain the observed precipitous decrease of defects available for microstructural evolution as defect production changes from predominately isolated defect pairs, i.e., during electron or light ion irradiations, to increasingly spatially close correlated formation in cascades, i.e., heavy ion or neutron irradiations. The most obvious cause is intra-cascade recombination. Averback et al. [10] have shown by resistivity measurements that the survival rate of defects during low temperature ion irradiation decreases significantly with increasing mass of the ion projectile, from close to the NRT value for protons to a saturation value of about 30% of the NRT value for intermediate and high mass ions. This would indicate that about 70% of the calculated defects annihilate by recombination during the cascade event for large cascades.

If one assumes that during irradiation at elevated temperatures additional intracascade recombination reduces the population of freely migrating defects to the values deduced from radiation-induced segregation or radiation-enhanced diffusion studies, one finds that almost no defects could escape from cascades producing more than about 5 defect pairs. [11] Along similar lines, Naundorf et al. [24,25] showed that single defect pairs created at a sufficient distance (>~ the spontaneous recombination radius) from any other defects in a cascade would account for the observed magnitude of freely migrating defect fraction. Neither of these approaches takes into account that a significant fraction of the defects surviving the cascade event are in the form of small defect clusters which contribute to the sink strength. [26,27] In the following we will include the effects of defect clusters on the freely migrating defect population.

Our general approach for discussing the freely-migrating-defect issue will be based on the following model description: Defects are produced in cascades. A cascade is characterized by the total number, N_o, of defects produced by collisional processes (NRT value). A fraction of defects, f_r, recombines (equal for vacancies, v, and interstitials, i); fractions f_{vcl} and f_{icl} of vacancies and interstitials, respectively, form immobile clusters; the remaining vacancies and interstitials, fractions f_v and f_i, enter the matrix as freely migrating defects. Note, only three of the five fractions f_v, f_i, f_r, f_{vcl} and f_{icl} are independent. The clusters formed provide sinks in addition to the conventional matrix sinks - dislocations, grain boundaries, voids and surfaces. Clusters grow by acquisition of freely migrating defects of the same type, and shrink by the precipitation of the opposite type of defects. During formation of a cascade, the central region corresponding to the 'molten' zone obliterates any pre-existing defect clusters (and free vacancies and interstitials) contained in the zone. The net number of obliterated defects, together with the fraction f_{vcl} of newly produced vacancies, reappears on resolidification of the zone as a single defect cluster. The new interstitial clusters created by the cascade (containing the fraction f_{icl} of N_o) are assumed to be formed outside of the molten zone.

With this set of assumptions, rate equations for the free defect concentrations can be set up and solved for steady state. Using the defect fluxes obtained, and the production and dissolution rates of clusters by cascades, the evolution of cluster size distributions can be integrated step-wise, recalculating the changing sink strength and the free defect fluxes in the steady-state approximation after each time step.

The rate equations for the time rate of change in the concentrations of point defects are somewhat modified from the usual form [28,29] :

$$dc_{i,v}/dt = k_{i,v} - Ac_ic_v - (v_{i,v}\, p_{i,v} + k_m)\, c_{i,v} \qquad (1)$$

where $c_{i,v}$ is the concentration, in atomic fraction, of freely migrating interstitials or vacancies (i,v), respectively, $k_{i,v}$ is the production rate of freely migrating interstitials or vacancies from all sources (cascades, thermal emission from sinks including clusters, and from clusters that shrink below their minimum stable, immobile size), A is the recombination rate constant containing the spontaneous recombination radius and the jump frequencies $v_{i,v}$ of defects, $p_{i,v}$ is the sink annihilation probability per defect jump at all sinks including immobile defect clusters, and k_m is the rate at which lattice sites are subjected to 'melting' in the central regions of cascades.

A few comments need to be made to emphasize the differences to the usual rate equations: the production rate of freely migrating interstitials is different from that of freely migrating vacancies because (i) of the different numbers of interstitials and vacancies lost to clusters in cascades and because (ii) of the greater excess rate of evaporation of vacancies from sinks, especially from small vacancy clusters. In fact, k_v can be greater or smaller than k_i depending on the prevailing cluster populations. For a given cluster population, $k_v - k_i$ may change sign as a function of temperature. The term involving elimination of freely migrating defects in the 'molten' zone, $k_m c_{i,v}$, limits the population of the slower moving defect species at low temperatures.

The rate of change in the concentration of immobile defect clusters of a given size, n, and species, e.g., vacancy clusters, can be expressed by

$$dm_{v,n}/dt = k_{cas,vn} + (j_{i,n+1} + e_{v,n+1})\, m_{v,n+1} + j_{v,n-1}\, m_{v,n-1} - (j_{i,n} + j_{v,n} + e_{v,n} + d_{cas,vn})\, m_{v,n} \tag{2}$$

Here, $m_{v,n}$ is the concentration of vacancy clusters containing n vacancies, $k_{cas,vn}$ is the rate of production of that size vacancy cluster directly by cascades, $(j_{i,n+1} + e_{v,n+1})$ is the sum of tne rates with which a cluster of the next larger size acquires interstitials and looses vacancies by evaporation (i.e., the rate at which a cluster of size n+1 shrinks to size n), $j_{v,n-}$

1 is the rate with which a cluster of the next smaller size acquires vacancies (interstitial evaporation is neglected), and $d_{cas,vn}$ is the rate at which a vacancy cluster of size n is dissolved by formation of new cascades. The assumption is made that clusters grow and shrink only via acquisition or loss of single point defects. The first three terms of the right hand side of eq.(2) represent the rate of gain, and the last term the rate of loss, of clusters of size n. For interstitial clusters an equation entirely equivalent to eq. (2) is valid.

As mentioned above, the set of equations for interstitial and for vacancy clusters of the form of eq. (2) is integrated using sufficiently small time steps with the coefficients in eq. (2) recalculated by solving eq. (1) for steady state defect concentrations applicable to the cluster distributions obtained after the previous time step The sink strengths and fluxes, $j_{i,n}$ and $j_{v,n}$, are calculated by the effective medium approach as discussed by Brailsford and Bullough [30], which also uses the steady-state approximation with regard to the mobile defects. The thermal emission rate of vacancies from clusters is determined with taking the difference in the energies of clusters of sizes n+1 and of n into account. The steady state approximation requires that the life times of defects are short compared to the lifetimes of immobile clusters.

4. Results from model calculations.

Calculations of the evolution of vacancy and interstitial cluster distributions have been performed for idealized irradiations in which all the defects are produced by a single cascade size. Certain approximations have been introduced to make the numerical calculations manageable without introducing a number of largely unknown physical quantities:

(i) Small defect clusters such as di-, tri-, ... vacancies and interstitials are mobile at temperatures of interest. Since the model assumes that the defect clusters in the distributions are immobile, we have introduced a lower size limit for immobile defect clusters. Those clusters which shrink below this size are assumed to decompose into mobile single vacancies or interstitials, which are included into the defect production terms, k_v and k_i.

(ii) Defect clusters that are larger than the size of the molten zone and, therefore, are unlikely to be dissolved by subsequent cascades, are removed from the calculated cluster size distributions. They are incorporated into the matrix sink strength as additional dislocation line length corresponding to the periphery of a circular prismatic dislocation loop containing the same number of defects.

(iii) For the purpose of calculation of the sink strength of defect clusters, all clusters are considered as spherical sinks of a size corresponding to the number of defects they contain.

(iv) The evaporation rate of vacancies from vacancy clusters is taken to be that corresponding to a spherical void containing the same number of vacancies.

(v) The fractions f_v and f_i of defects released from the cascades into the matrix were scaled for the cascade size by an exponential decay from 1 for isolated defect pairs, to the limiting values, f_{vo} and f_{io}, for large cascade sizes (see Table 1). Similarly, the fraction, f_r, of defect pairs lost by recombination within the cascade was scaled to vary from zero to approach the limiting value, f_{ro}, for large cascade sizes with the same decay length.

The reference values for the cascade characteristics are listed in Table 1. They were chosen to be consistent with the values obtained by Heinisch from binary collision calculations (MARLOW) with subsequent "short-time annealing". [31,32]]The factor connecting the size of the molten zone to the number of defect pairs (or to the damage energy) is consistent with molecular dynamics calculations by T. Diaz de la Rubia and M.W. Guinan. [13] The physical quantities, such as vacancy and interstitial migration and formation energies, for the sample calculations were chosen to be representative of nickel [33, 34] and are listed in Table 2.

Most calculations were performed with the reference values of Table 1 for a cascade size $N_o = 200$ defect pairs, corresponding to a PKA energy of about 15 keV. This cascade size was chosen to be representative of the average subcascade size in high energy primary recoils. All results of calculations presented here were obtained for a NRT displacement rate of 10^{-3} dpa. Results for other displacement rates are similar when compared at the same dose.

The development of the defect cluster distributions are best discussed separately for the two temperature regimes distinguished by whether interstitials or vacancies are released in excess from cascades and from defect clusters into the matrix. The net release rates of defects (given in atom fraction/dpa) are shown in Fig. 1 as a function of the total dose for several temperatures. After a short transient, a few tenth of a dpa, the net release rate tends toward a quasi steady state value which is positive (excess interstitials) below about 620 K and is negative (excess vacancies) at intermediate temperatures. At temperatures above 650 K, the approach to a stationary value becomes slow.

In the lower temperature regime, interstitial release dominates because of the larger number of interstitials than vacancies released from cascades. As a consequence, interstitial clusters tend to grow and vacancy clusters tend to shrink. The former is illustrated in Fig. 2, which shows that the interstitial cluster distribution develops a long tail toward large sizes with increasing dose. Note that the ordinate in Fig. 2 is logarithmic: the vast majority of clusters remain small, and their distribution is established at small doses. Large loops which should be observable by TEM develop slower and at much lower concentrations. The vacancy cluster distributions drop precipitously for larger sizes and the drop becomes somewhat more pronounced at larger doses. The model results seem consistent with microstructural observations at the low end of and below the void swelling regime.

As temperature increases, the rate at which vacancies are released by "evaporation" from small vacancy clusters increases, and ultimately the total rate of vacancy release, k_v, exceeds that of interstitials, k_i, in the upper temperature regime, see Fig. 1. As illustrated in Fig. 3, the development of the interstitial and vacancy cluster distributions inverts qualitatively as expected: The vacancy cluster distribution develops now a long, low-concentration tail toward large sizes, whereas the interstitial cluster distribution becomes sharply curtailed for larger sizes. Provided that at least part of the larger vacancy clusters have remained three-dimensional clusters (they are beyond critical size as void embryos), void swelling should commence even in the absence dislocation bias which has been neglected in the present treatment. The cause for this swelling is the "production bias" suggested by Singh and Woo.

When the temperature is increased further by a moderate amount, the development of the vacancy cluster distribution is somewhat more complex as shown in Fig. 4. The development of a minimum in the vacancy cluster distribution is caused by a decrease in the number of vacancies that cluster upon the solidification of the molten zone, in conjunction with the faster loss rate of the smaller vacancy clusters at creation. The decrease of the number of vacancies in the clusters at formation is a consequence of the increasing number of interstitials dissolved from clusters in the molten zone, relative to that of vacancies, as the dose increases. The model indicates that vacancy clusters which have grown to sufficiently large sizes at the beginning of the irradiation continue to grow because of the excess rate of vacancies released to the matrix. Thus, the model yields early void nucleation that ceases at a relatively small dose.

The interstitial and the vacancy cluster distributions that develop during irradiation to 5 dpa at several temperatures are compared in Fig. 5 and Fig. 6, respectively. The distributions reflect the features discussed above: Interstitial cluster distributions extend to large sizes at low temperatures, but become increasingly restricted to smaller sizes as vacancy release from clusters gains in importance at higher temperatures. Conversely, vacancy clusters are confined to small sizes at temperatures below swelling; as vacancy evaporation from small clusters becomes important, vacancy clusters grow to large sizes. At still higher temperatures, small vacancy clusters become extinct at relatively low doses; however, clusters that have grown to sufficiently large sizes in the early stages of irradiation continue to grow.

The contributions of the free defect fluxes to diffusion are illustrated in Fig. 7. It should be emphasized that displacement mixing and diffusional processes during the cascade process, such as recombination and defect clustering, are not included in the diffusion coefficients shown. In Fig. 7 the total diffusion coefficient by thermal defect motion, and the individual contributions of vacancies and interstitials, are plotted for 500 K and 660 K. The diffusion coefficients decrease rather quickly, by more than an order of magnitude, with dose as the defect cluster populations build up and then remain relatively constant. As expected, at the lower temperature at which there is an excess flux of interstitials, interstitials contribute more to atom transport than freely migrating vacancies. At the higher temperature at which small vacancy clusters evaporate quickly, vacancies contribute more than interstitials to atom transport. Although the proposed model is qualitatively consistent with the experimentally observed low efficiency of defects produced by heavy ions in enhancing diffusion or inducing segregation, quantitatively the values predicted for the enhanced diffusion are lower than those extracted from experiment. The major reason for this is probably an overestimate, by the model, of the population of small interstitial clusters which provide a major contribution to the sink strength. Increasingly larger interstitial clusters will become mobile with increasing temperature. Cluster mobility should lead to significant losses of small clusters by reactions with other clusters, network dislocations and grain boundaries.

Model calculations have also been performed other cascade sizes and with different values of the cascade parameters, e.g., the fractions of recombining and escaping defects, sizes of the molten zones and sizes of the minimum immobile defect clusters (below which defect clusters are assumed to decompose spontaneously). The results are surprisingly similar in characteristics, although the numerical values change moderately for reasonable ranges of the cascade parameters.

Summary.

The direct formation of defect clusters in cascades has important consequences on the evolution of the microstructure during irradiation. A simple model invoking the formation of both vacancy and interstitial clusters directly in cascades can account for low efficiencies of production of free defects in cascade producing irradiations, and describe the evolution of damage microstructures semi-quantitatively at low and intermediate temperatures. It indicates that interstitial and vacancy cluster distributions develop quickly as a function of dose. Sizes near those of formation are highly dominant in the cluster populations and, therefore, in their contributions to the total sink strength. At low temperatures, the flux of free interstitials exceeds that of vacancies. As a consequence, a low concentration tail of large interstitial dislocation loops develops whereas the vacancy cluster distribution remains confined to small sizes.

The model accounts for a transition, with increasing temperature, to a void swelling regime when the evaporation of vacancies from small vacancy clusters in the distribution becomes increasingly fast and, hence, the flux of freely migrating vacancies becomes larger than that of interstitials. In this situation, the vacancy cluster distribution develops a long, low concentration tail toward large sizes. Vacancy clusters that have remained void-embryo like can grow beyond the critical void size and initiate void swelling. The interstitial cluster distribution remains confined to small sizes because of the excess flux of vacancies. As the temperature is increased further, the model predicts that vacancy clusters evaporate so quickly that their concentration becomes negligible and no void nucleation takes place. A significant concentration of small interstitial clusters would persist at high temperatures as a consequence of the assumption in the model that interstitial clusters above a fixed minimum size are considered immobile.

The model does not reflect the high temperature behavior (peak swelling and above) of the microstructural development correctly. The most likely reason for this failure is the assumption of the immobility of larger interstitial clusters and the replacement of the mobility of smaller interstitial clusters by their decomposition into single interstitials. The use of a temperature dependent minimum size of immobile interstitial clusters could improve the results of the model calculations. However, as shown by Trinkaus, the piece-wise one-dimensional diffusion (between spontaneous changes in Burgers vector) of small interstitial clusters by glide should lead to strongly preferred removal of interstitial clusters at extended sinks, such as grain boundaries and network dislocations, relative to localized sinks, i.e., defect clusters. This preferred elimination of interstitial clusters is not reflected in the model since the assumed decomposition leads to three-dimensional diffusion of single interstitials. This short coming of the model is expected to be become increasingly severe at higher temperatures and with the attendend lower vacancy cluster densities. In order to make the model more quantitative, the temperature and size dependent mobility of small interstitial clusters needs to be incorporated. It should be noted that the model can be adapted rather straight forward to incorporate the entire primary knock-on spectrum for a given radiation environment.

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Figure captions.

Fig. 1. Net rate of release of defects into defects into the matrix as function of dose for a number of irradiation temperatures. The excess interstitial flux is proportional to the net rate of release at steady state.

Fig. 2. Development of defect cluster distribution at 600 K. The doses are indicated at the curves in dpa.

Fig. 3. Development of defect cluster distributions at 650 K. The doses are indicated at the curves in dpa.

Fig. 4. Development of the vacancy cluster disrtibution at 660 K.

Fig. 5. Interstitial cluster distributions after irradiation to 5 dpa at several temperatures.

Fig. 6. Vacancy cluster distributions after irradiation to 5 dpa at several temperatures.

Fig. 7. Diffusion coefficient from thermal motion of all defects (total), of vacancies and of interstitials. Open symbols: 500 K, solid symbols: 660 K.

Table 1. Characteristics of reference cascades:

T	= 15 keV	PKA energy
N_o	= 200	number of defect pairs (NRT value)
f_{ro}	= 0.77	fraction of N_o lost by recombination
f_{vclo}	= 0.22	fraction of vacancies in clusters
f_{iclo}	= 0.19	fraction of interstitials in clusters
f_{vo}	= 0.01	fraction of freely migrating vacancies
f_{io}	= 0.04	fraction of freely migrating interstitials
f_{mo}	= 15	size of molten zone : N_o*f_m = 3000 sites
n_d	= 40	decay size (size at which fractions are approch asymptotic value to within 1/e)

Table 2. Values for defect parameters in Ni used for model calcul;ations.

Vacancy	formation enthalpy	E_{vf}	=	1.58 eV
	formation entropy	S_{vf}	=	1.5 k
	migration energy	E_{vm}	=	1.30 eV
	attempt frequency	n_o	=	$1.98\ 10^{15}\ s^{-1}$
Interstitial	formation enthalpy	E_{vf}	=	1.58 eV
	formation entropy	S_{vf}	=	1.5 k
	migration energy	E_{vm}	=	1.30 eV
	attempt frequency	n_o	=	$4.23\ 10^{12}\ s^{-1}$

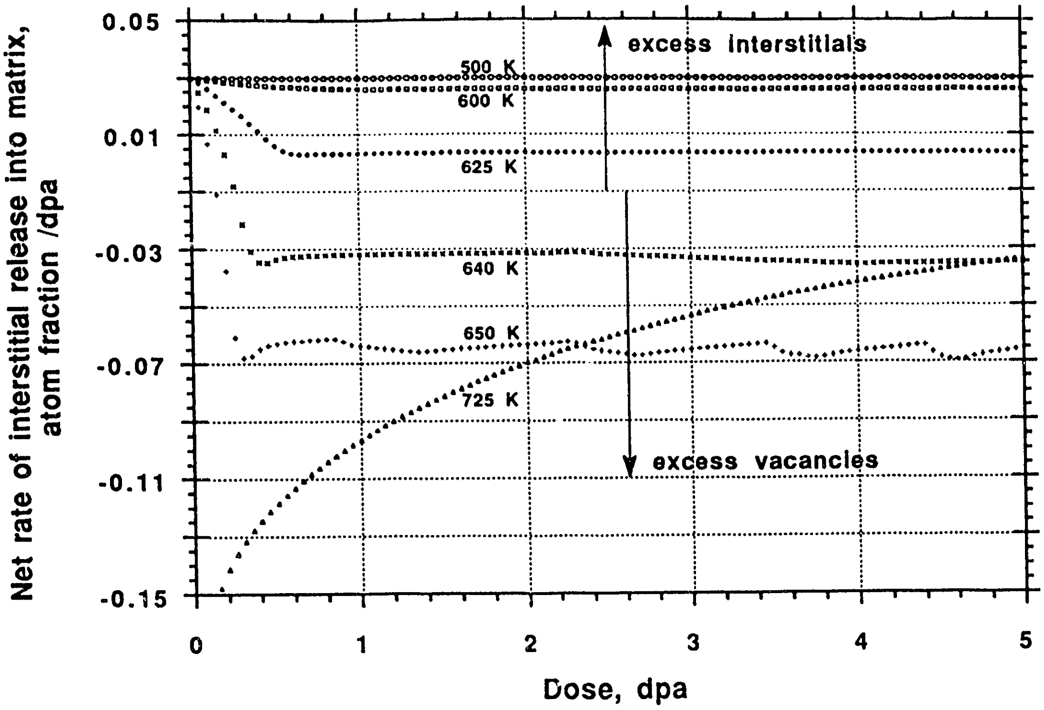

Fig. 1. Net rate of release of defects into defects into the matrix as function of dose for a number of irradiation temperatures. The excess interstitial flux is proportional to the net rate of release at steady state.

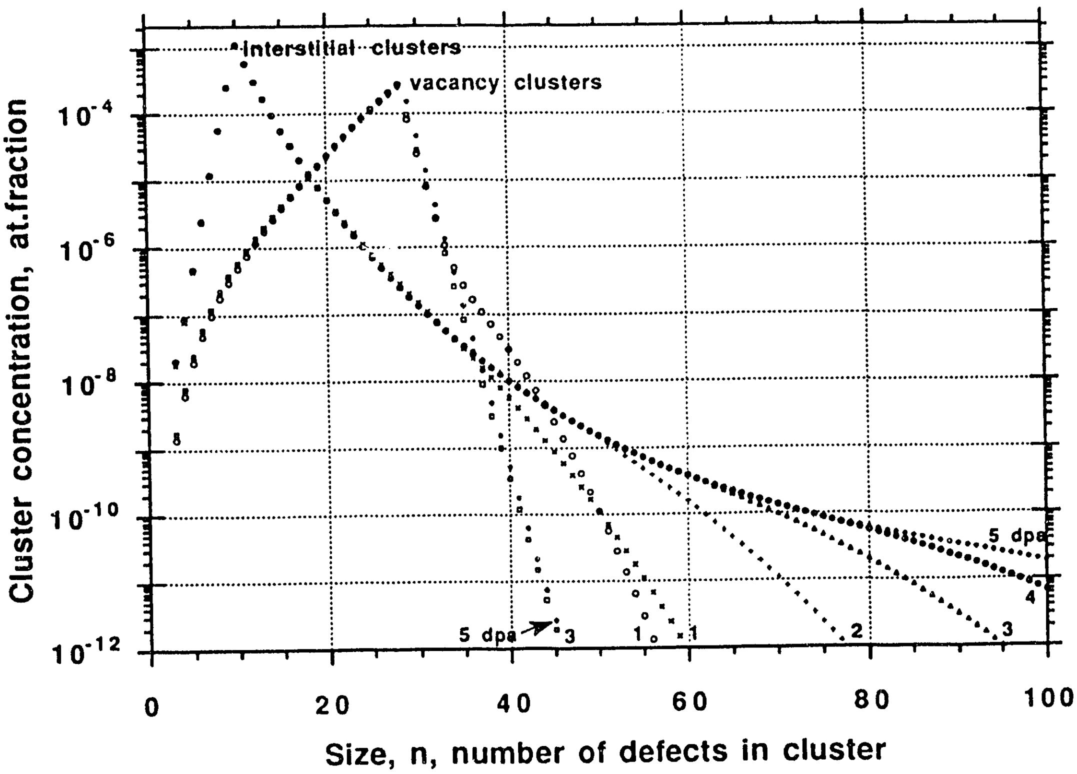

Fig. 2. Development of defect cluster distribution at 600 K. The doses are indicated at the curves in dpa.

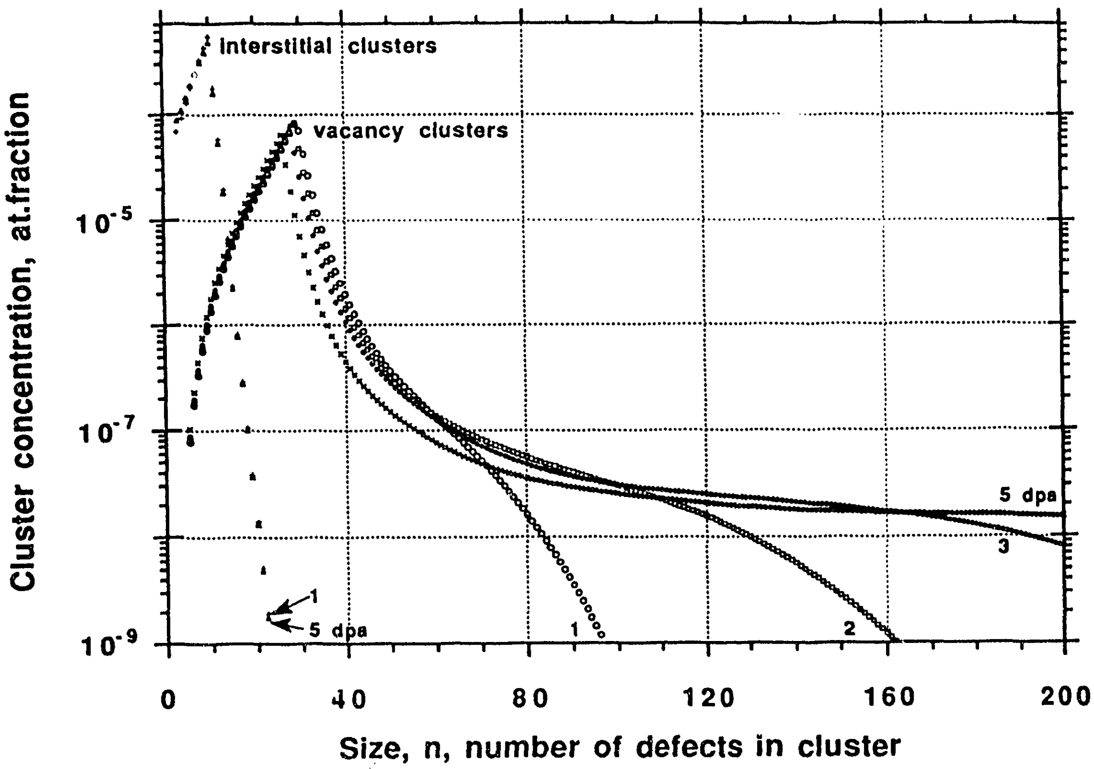

Fig. 3. Development of defect cluster distributions at 650 K. The doses are indicated at the curves in dpa.

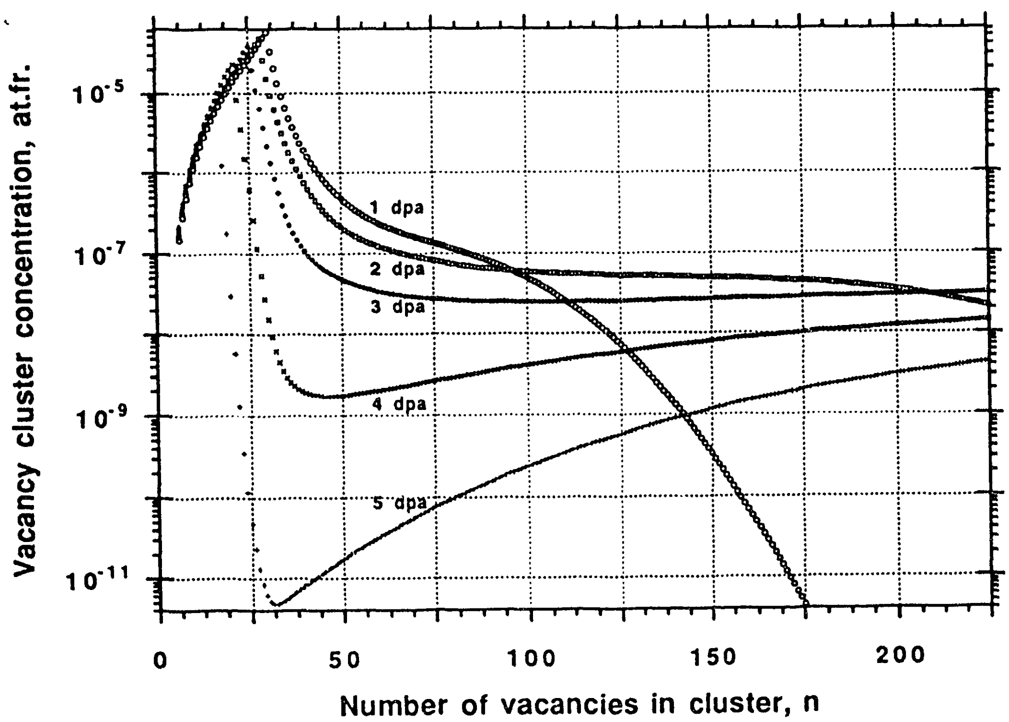

Fig. 4. Development of the vacancy cluster disrtibution at 660 K.

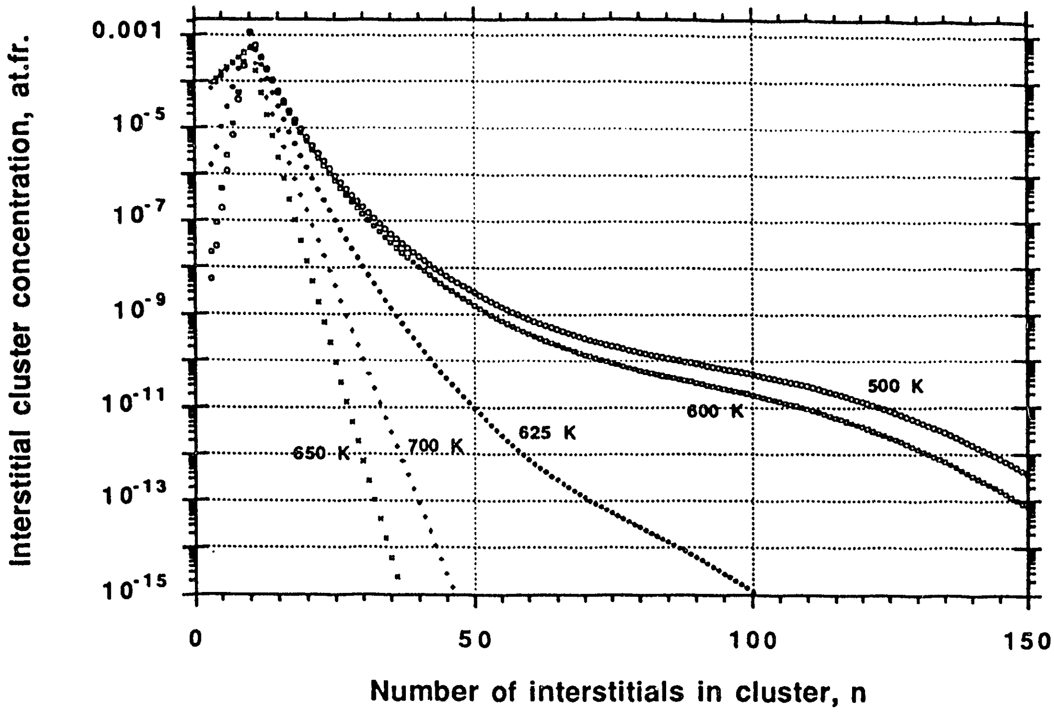

Fig. 5. Interstitial cluster distributions after irradiation to 5 dpa at several temperatures.

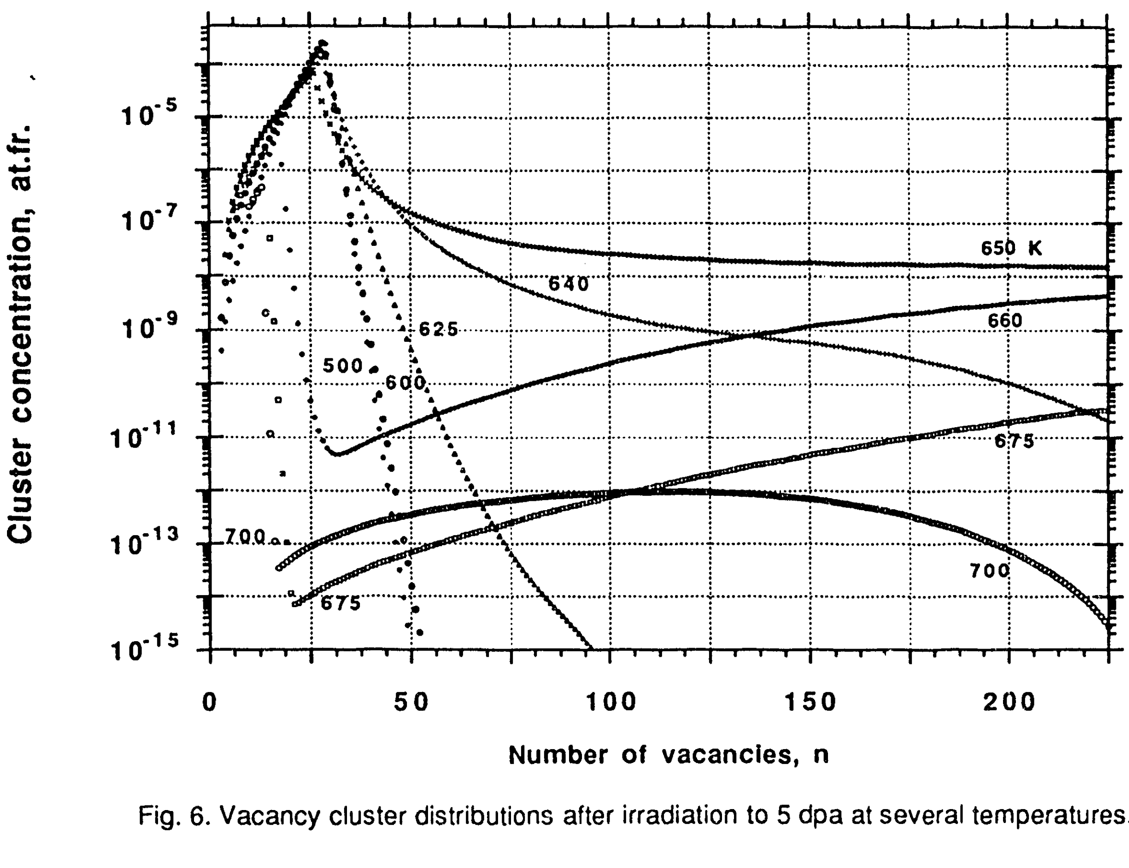

Fig. 6. Vacancy cluster distributions after irradiation to 5 dpa at several temperatures.

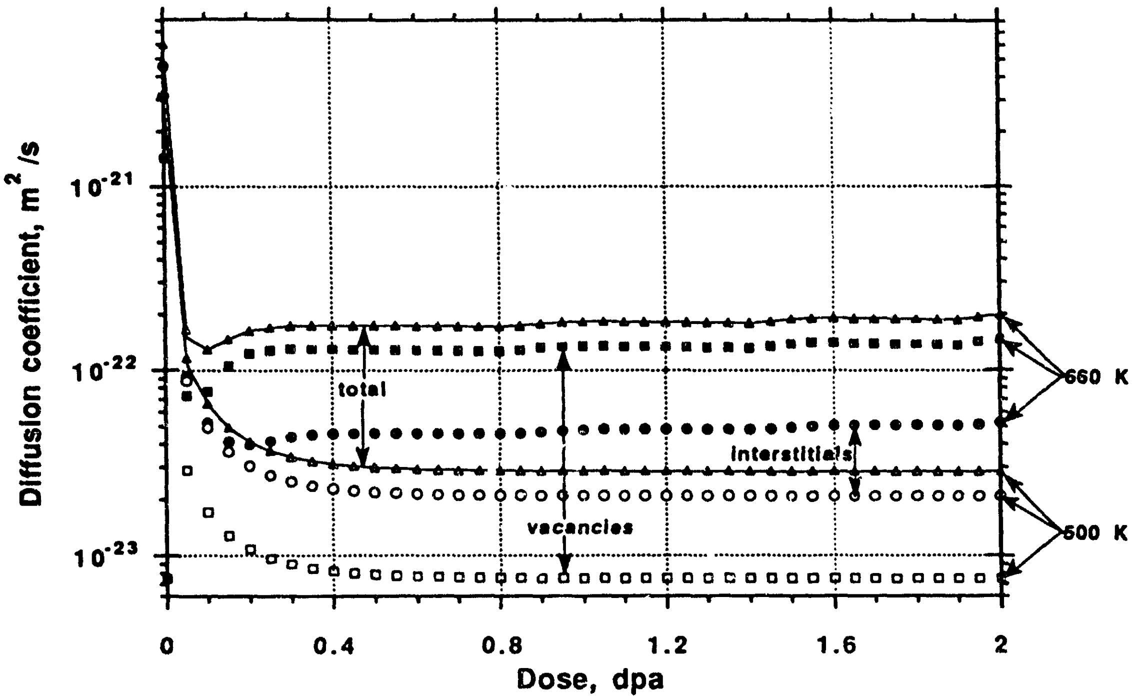

Fig. 7. Diffusion coefficient from thermal motion of all defects (total), of vacancies and of interstitials. Open symbols: 500 K, solid symbols: 660 K.

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