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Impacts of damage production and accumulation on materials performance in irradiation environment

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Abstract

Aspects of generation of atomic displacements and surviving defect fractions and their sensitivity to recoil energy have been considered. These considerations show that the nature of the primary damage production plays a vital role in the evolution of the damage accumulation as well as mechanical properties. The problem of damage accumulation on a global scale still remains a theoretical challenge! A substantial amount of theoretical effort is required to develop a realistic model to describe the damage accumulation at low temperatures (<stage V) and to establish a reliable relationship between the damage accumulation, irradiation hardening and the loss of ductility. The recently developed production bias model together with one-dimensional glide of interstitial clusters produced in the cascades has been shown to describe the damage accumulation at temperatures above stage V for pure metals and needs to be extended to temperatures below stage V and to materials of practical interests. This requires, however, that the information regarding the effects of alloying elements and impurity atoms on the nature of the primary damage state are available from molecular dynamics and kinetic Monte Carlo type of simulations.

1. Introduction

It is well established that irradiation with energetic particles causes substantial changes in physical and mechanical properties of metals and alloys. From the technological point of view, this has been a matter of serious concern since these changes are likely to affect the performance and lifetime of materials used in different components of a fission or fusion reactor. This concern has provided the driving force for a large number of experimental investigations carried out over the years starting already in 1950s (e.g. [1]). Irradiation experiments demonstrated very rapidly that materials exposed to fission neutrons at relatively low temperatures (i.e. $<0.3T_m$, where T_m is the melting temperature) harden significantly already at low doses and suffer from a drastic reduction in their ductility. These phenomena became commonly known as irradiation hardening and irradiation-induced embrittlement and are still being studied intensively (see later).

Another major effect of neutron irradiation is the accumulation of vacancies in the form of voids [2] at temperatures above $0.3T_m$ particularly in fcc metals and alloys. The phenomenon became known as void swelling and over the years has been studied extensively both experimentally and theoretically. During irradiation with neutrons another complication arises since neutrons produce not only displacement damage in the lattice but also generate a variety of solid and gaseous impurity atoms via nuclear reactions. In particular, the generation of helium is a matter of concern since helium atoms are practically insoluble in metals and alloys and can be potent agent in enhancing void formation specially at high temperatures. Furthermore, helium atoms may segregate at grain boundaries in the form of bubbles and may cause grain boundary embrittlement.

Now, the question arises as to how and why the irradiation with energetic particles induces such profound changes in the properties of materials particularly when the irradiation-induced self-interstitial atoms (SIAs) and vacancies are "matter", "anti-matter" type of defects

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which can annihilate each other and repair the lattice! Before addressing these questions, it would be instructive, however, to consider a comprehensive picture of the events and processes controlling the microstructural evolution and thus the properties of materials under irradiation. This is illustrated schematically in Fig. 1. It can be easily seen that the response of a given material (in terms of hardening, swelling, embrittlement etc.) to irradiation under a given set of conditions is going to depend on the nature and level of damage accumulation in various forms (e.g. defect clusters, loops, SFTs, dislocation decoration, dislocation density, voids, bubbles at grain boundaries, etc., etc.). The damage accumulation itself, on the other hand, is likely to depend on the form in which the primary damage is produced. At low recoil energy, for instance, the damage will occur entirely in the form of isolated Frenkel pairs. Furthermore, at this low recoil energy neither clusters of defects nor transmutational impurities will be produced. The damage accumulation under these conditions will be significantly different from that under the condition of very high recoil energy when the primary damage will be dominated by the production of intracascade clusters. The damage accumulation will be further complicated under those irradiation conditions where nuclear reactions are likely to produce significant amounts of transmutational impurities (solid and gaseous).

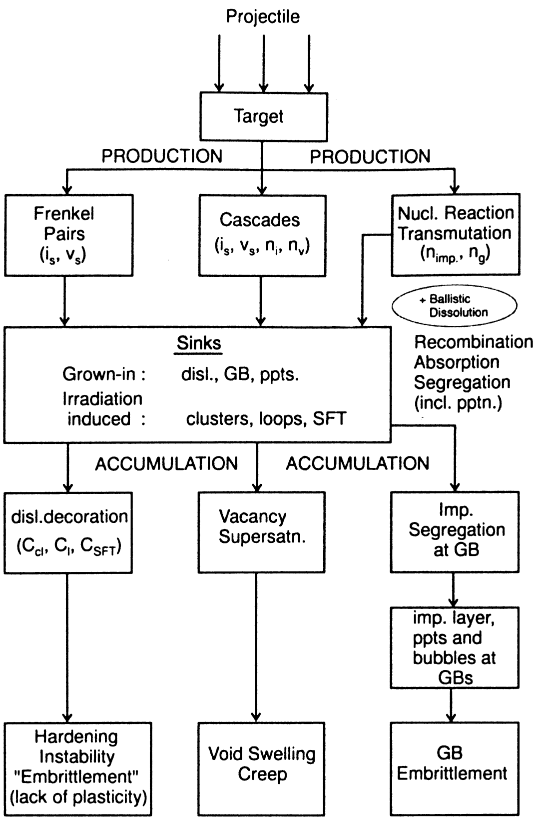

Fig. 1. Schematic illustration of damage production, sink interaction and accumulation responsible for irradiation effects such as low temperature hardening and "embrittlement, void swelling and grain boundary embrittlement" at higher temperatures.

Thus, it can be seen that in order to discuss and evaluate the performance and lifetime of materials exposed to different irradiation conditions, it is necessary to understand the nature of the primary damage production and its role in the process of damage accumulation and microstructural evolution. In order to facilitate this discussion we first consider the problem of generation of atomic displacements and surviving defects and their clusters in Section 2. This is followed by physical considerations of fundamental issues involved in the temporal evolution of defect accumulation on a global scale (Section 3). Both theoretical and experimental aspects of the damage accumulation in the form of voids are also briefly discussed in Section 3. The impact of irradiation-induced defects and their clusters on the deformation behaviour of metals and alloys is treated in Section 4. A brief summary and main conclusions are given in Section 5.

2. Damage production: atomic displacements and lattice defects

The main elementary interaction during irradiation is the elastic collision between the projectile particles and the atoms of the solid target. During this event, when the recoil energy transferred to a target atom exceeds the displacement threshold energy, the struck atom is displaced from its original site, creating a vacancy-interstitial pair (commonly known as Frenkel pair). Historically, Kinchin and Pease [3] were first to estimate the number of displaced atoms (or Frenkel pairs) in terms of the amount of energy transferred to the primary knock-on atom (PKA) and the displacement threshold energy for a given solid. This model was modified by Norgett, Robinson and Torrence (NRT) [4] who took into account the loss of energy to electron excitation.

It should be noted that the internationally accepted unit of displacement damage dpa (displacement per atom) is based on the NRT model. It is important to note, however, that the number of surviving defects, which is the crucial parameter for the microstructural evolution, decreases very rapidly with increasing recoil energy (see Section 2.1). During the neutron irradiation of copper, for instance, at void swelling temperatures the number of surviving defects may be as low as only 10% of the displacements generated in the collisional phase [5]. A critical review on this subject can be found in [6]. Details of the energy transfer and the basic mechanisms of displacement generation are dealt with in a paper in the present proceedings by Ghoniem [7] and therefore will not be repeated here.

2.1. Displacement cascades

When the energy transferred to PKAs by high energy projectile particles becomes substantially higher than the displacement threshold energy, instead of a single isolated displacement, a hierarchy of displacements are produced in a relatively small volume and in a very short time (less than a peco-second). The qualitative concept of the production of such *cascades* of displacements was introduced already in 1950s by Brinkman [8] and Seeger [9]. In 1960s, Linhard and co-workers [10,11] laid down the foundation of collision physics describing the production of the nascent damage state including cascades and subcascades of displacements. However, the results of their elementary theory of displacement cascades could not provide a detailed description of the temporal and spatial evolution of the displacements. Furthermore, the collision physics description of a displacement cascade could not be extended to determine the amount, morphology and spatial distribution of lattice defects and their clusters surviving at the end of the cooling down (i.e. thermal spike) phase of the cascade.

Over the years, attempts have been made, however, to solve this problem by using molecular dynamics (MD) as a tool for computer simulation of displacement cascades. Although the MD simulation technique was pioneered already in 1960s [12,13], it is only recently that the availability of more reliable interatomic potentials and more powerful computer has made it possible to simulate displacement cascades by recoils with energies as high as 40 keV [14]. Within the last decade, a large number of simulation studies of displacement cascades have been carried out in various metals (see [15–17] for reviews). It should be mentioned, however, that the largest number of simulation experiments have been targeted on copper [18,19] and bcc iron [14,15,20–22].

Undoubtedly, the MD simulations of displacement cascades provide the most valuable knowledge about the surviving defects, their nature, disposition and morphology. However, these simulations and the subsequent analyses are extremely time consuming and even with the present computing speed and capacity it does not seem very practical to simulate displacement cascades with as high recoil energies as are likely to be produced by 14 MeV neutrons. As can be seen in Fig. 2 [23], the primary recoils generated by fusion neutrons will have energy in the range of 100s of keV and will create damage in the form of multiple cascades and subcascades. Such high energy events can be simulated, on the other hand, using binary collision approximation (BCA) code MARLOWE [24] (see [25] for a review). It should be emphasized, however, that this type of simulations provide information regarding the density and disposition of the displacements and the number and morphology of subcascades produced at the end of the collisional phase.

Thus, the simulations using MARLOWE code on its own cannot yield information on the details of the lattice defects surviving at the end of the thermal spike phase. However, in order to extract at least some qualitative information regarding the production of lattice defects from these simulations, a Monte Carlo annealing simulation code ALSOME was developed [26] for quenching and short term annealing of the cascades produced by MARLOWE. Using this combination of MARLOWE and the stochastic annealing code ALSOME,

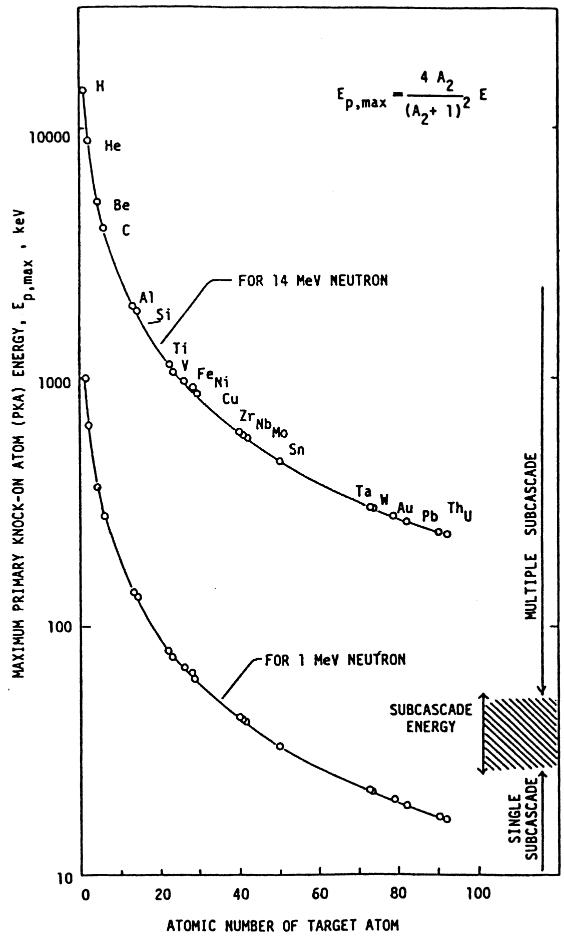

Fig. 2. Variation of maximum recoil energies produced by 1 and 14 MeV neutrons with atomic number of the target materials [23] with atomic mass A_2 for a number of elements. Note that the fusion (14 MeV) neutrons will generate PKAs with much higher energy than those generated by the fission (1 MeV) neutrons. This difference would lead to significant differences in the production of the primary damage state.

various aspects of cascade and subcascade production at high recoil energies have been studied systematically in fcc, bcc and hcp metals as a function of recoil energy [27]. Parameters such as the average local vacancy concentration in the cascade core and the average number of subcascades produced per PKA were determined as a function of the damage energy. These investigations led to two important conclusions. First, the average local vacancy density in the core of a cascade increases with increasing atomic number. Second, the average number of subcascades per PKA (or cascade) increases linearly with increasing recoil damage energy (Fig. 3). At a given recoil damage energy, the number of subcascades in different metals decrease with increasing atomic number. Thus, binary collision simulations can be used to provide a statistically meaningful description of the initial cascade morphology as a function of recoil energy as well as materials parameters.

2.2. *Important features of defects and their clusters in displacement cascades*

As indicated in the preceding section, by now a large number of cascade events have been studied using the MD simulation technique. However, all these studies have described the intracascade events in *isolated* single cascades. It remains uncertain, therefore, as to whether or not the basic nature of the intracascade evolution of defects and their clusters may be significantly influenced by the production of other cascades in the medium. There are still uncertainties regarding the accuracy and appropriateness of the interatomic potentials used in MD simulations. The time-scale used in these simulations is rather short (of the order of peco-seconds) and may not allow a complete relaxation of the defects and their configuration. In spite of these uncertainties, however, the MD simulation results have demonstrated some general trends of behaviour which are also consistent with experimental results (see later). These trends must be taken seriously since some of these unique features of defect production lead to fundamental changes in the consideration of damage accumulation under cascade damage conditions (see [6] for a review).

Based on the published MD results [14–22], the following features of defect production in displacement cascades can be identified:

(i) At PKA energies of ~0.5 keV and above the damage production occurs in the form of displacement cascades.

(ii) Intracascade recombination of SIAs and vacancies during the thermal spike phase leads to a decrease in the fraction of surviving defects (i.e. the damage efficiency) with increasing PKA energy. The decrease is very rapid up to a PKA energy of about 5 keV and in copper reaches a value of ~20% of the NRT value at the PKA energy of ~10 keV at 100 K. Beyond this energy level, the decrease in the damage efficiency is not very large (see Fig. 4) [28].

(iii) Intracascade clustering of both SIAs and vacancies occurs during the cooling-down phase of the cascades.

(iv) The fraction of SIAs surviving in the form of clusters increases with increasing PKA energy; this fraction is higher in copper than in α-iron (see Fig. 5) [17].

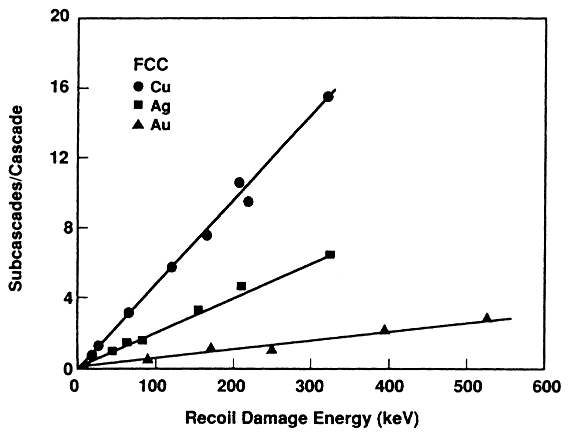

Fig. 3. Effect of recoil damage energy on the number of subcascade production per cascade in copper (●), silver (■) and gold (▲) calculated using the BCA code MARLOWE [27]. Note that the production of the primary damage state will be sensitive not only to the recoil energy but also to the atomic mass density of the target material.

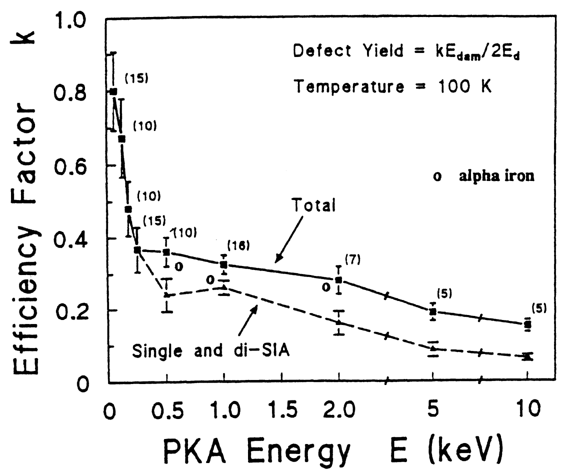

Fig. 4. Variation of the NRT damage efficiency factor for defect production as a function of PKA energy obtained from MD simulations [28] in copper and iron. Note the decrease in the efficiency of defect production with increasing PKA energy.

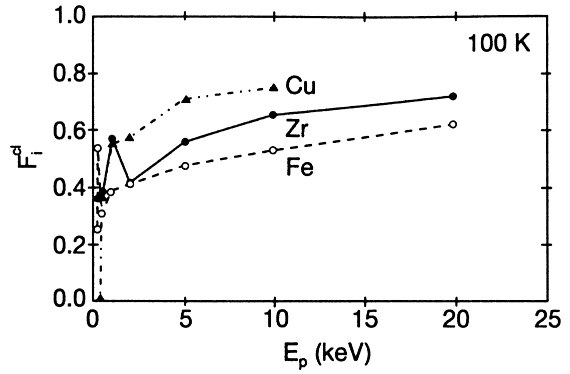

Fig. 5. Variation of the interstitial clustering fraction, F_i^{cl}, obtained from MD simulations as a function of cascade energy for copper, α-iron and zirconium at 100 K [17]. Note that there is a substantial difference in the intracascade clustering behaviour of SIAs between copper and α-iron.

(v) The intracascade clustering occurs in a segregated form such that the vacancy cluster is formed at the cascade centre, whereas the clusters of SIAs are formed at the periphery of the cascade.

(vi) Small clusters of SIAs produced in the cascade during the thermal spike phase are glissile.

Most of these features described above have been found to be consistent with experimental results. The decrease in the damage efficiency with increasing recoil energy observed in MD simulations is in reasonably good agreement with the values determined from electrical resistivity measurements made on several metals during irradiation at ∼4 K (see Fig. 2 in Ref. [6]). The presence of clusters of SIAs and vacancies in metals irradiated at ∼4 K was determined by diffuse X-ray scattering experiments [29–31]. Although there is no direct experimental evidence demonstrating the glide of small SIA clusters, indirect evidence can be deduced from experimental results on the evolution of SIA clusters in annealing stage II after low temperature electron [32,33] and neutron irradiation [31,33,34], the formation of rafts of loops [35,36] and decoration of grown-in dislocations by small SIA loops [37].

The brief review presented in this section is summarized in Fig. 6 by schematically illustrating various surviving defect fractions generated under single and multiple atomic displacement (cascade) conditions. It can be easily seen that the nature of defect generation under cascade damage conditions is dramatically different and immensely more complicated than that under single displacement (Frenkel pair) production condition. It is important to make the following observations regarding the nature and the magnitude of the surviving defect fractions shown in Fig. 6. First, since the level of PKA energy depends not only on the energy of the projectile particles but also on their mass and the mass of the target atoms, the details of the surviving defect fractions are expected to be sensitive to the irradiation environment as well as the target material. The second point concerns the influence of alloying elements on defect production in cascades since the presence of alloying elements may affect the collisional phase as well as the size and the lifetime of the thermal spike [17,38]. This in turn may affect the magnitude of various fractions of the surviving defects. Finally, it should be added that even the crystal structure and electronic property of metals may have influence on the surviving defect fraction [21,39]. Recent MD simulations suggest that practically all SIA clusters produced in cascades in bcc iron are likely to be glissile [40].

3. Damage accumulation

The accumulation of "matter", "antimatter" types of defects produced during irradiation, which in principle should annihilate each other, has remained an intriguing

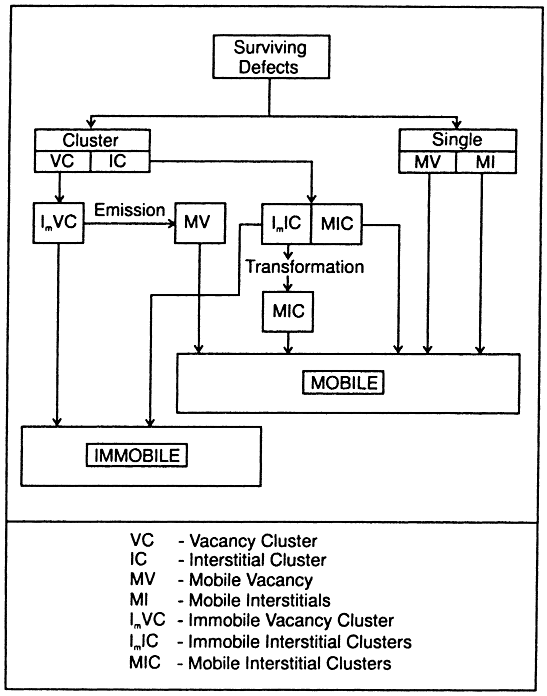

Fig. 6. Schematic illustration of various fractions of surviving defects produced under conditions of Frenkel pair (i.e. single defects) production and multidisplacement cascade production where both single defects and defect clusters are produced. Note that at temperatures above stage V, the fraction of the mobile SIA clusters is made up of mobile clusters produced directly in the cascades and those that result from the transformation of immobile clusters into the mobile ones.

problem for almost half a century. This appears to be primarily because of the fact that the process of defect production itself, particularly under cascade damage conditions, is a very complex one. As discussed in the preceding section, the SIAs and vacancies are spatially segregated already at the production stage and are formed as clusters with different properties. Consequently, they cannot recombine and annihilate each other. The facts that the SIA clusters are glissile and are considerable more stable against the thermal decomposition than the vacancy clusters give rise to serious complications (see later) in formulating an appropriate description of the defect accumulation kinetics. However, in recent years, some significant progress have been made in this area. The main ideas driving these new considerations and their main achievements will be briefly described in this section.

3.1. General considerations

Let us first consider the general aspects of the build-up of surviving defects and their clusters (see Fig. 6) in the form of a global microstructure which can be characterized experimentally either during the irradiation or in the post-irradiation state. The evolution of the irradiation-induced microstructure can, of course, be studied theoretically within the framework of homogeneous reaction kinetics using chemical rate equations. In addition, Kinetic Monte Carlo (KMC) type of computer simulations can be employed in a large enough volume with periodic boundaries to determine the temporal evolution of the microstructure during continuous irradiation as in a real irradiation experiment. Clearly, both the analytical calculations and the KMC type of simulations will have to depend on the information regarding the defect production parameters obtained from the MD simulations of single, isolated cascades. This may become a source of uncertainties in the results of both analytical calculations and KMC simulations if the defect production parameters in the global situation were found to be significantly different from that obtained for single cascades.

Finally, it should be mentioned that the treatment of damage accumulation under cascade damage conditions must also include an appropriate consideration of the difference in the thermal stability between clusters of SIAs and vacancies. At temperatures above the recovery stage V vacancies will begin to evaporate from the clusters and will escape into the medium contributing to the vacancy supersaturation. Using a diffusion calculation, it was shown by Singh and Foreman [41] that about 80% of the vacancies evaporating from a vacancy cluster in the centre of a cascade surrounded by SIA clusters at the cascade periphery would escape the cascade volume. This result has been recently confirmed by KMC simulations [15,42]. The SIA clusters, on the other hand, are thermally stable at void swelling temperatures [43].

3.2. Damage accumulation: A theoretical challenge!

At low recoil energies when the damage energy is just above the displacement threshold (e.g. during 1 MeV electron irradiation), the displacement damage occurs in the form of single, isolated SIAs and vacancies. For such simple irradiation cases, the damage accumulation can be treated successfully within the framework of mean-field theory using chemical rate equations (i.e. standard rate theory, SRT) (e.g. [44,45]). It has been shown recently, for instance, that the damage accumulation in pure copper irradiated with 2.5 MeV electrons calculated using SRT is in very good agreement with the experimental results [46,47]. At higher recoil energies, however, the damage production in the form of cascades introduces a number of complications (Fig. 6), as described in the preceding section. Under these conditions, a reliable and realistic calculation of the build-up of the defect populations in the form of a global microstructure containing loops, stacking fault tetrahedral (SFTs) and cavities is a real theoretical challenge. It should be pointed out here that even the models such as the BEK [48] and the so-called "composite" model [49] (which is essentially the same as the BEK) are grossly inadequate to describe the damage accumulation under cascade damage conditions (see [50] for a detailed analysis).

The recognition of the importance of intracascade clustering of SIAs and vacancies, of the difference in thermal stability between SIA and vacancy clusters and of the possibility that the fraction of SIAs contained in the SIA clusters produced in the cascades may be different from that of the vacancies in the vacancy clusters led Woo and Singh [51,52] to propose the concept of production bias. The production bias model (PBM) was shown to provide a driving force for void swelling in the steady-state and to explain the temperature dependence of the steady-state void swelling. Subsequently, the evolution of void swelling in the transient regime was calculated in terms of PBM by Singh and Foreman [41]. The results demonstrated that the PBM is indeed capable of predicting the experimentally measured swelling behaviour of neutron irradiated copper at 250°C. These calculations also showed (a) that in order to explain the observed swelling at higher doses it was necessary to assume that 15% of the SIA clusters produced in the cascades escaped to sinks other than voids by one-dimensional glide and (b) that without such losses, the build-up of SIA clusters will continue to increase to unrealistically high values. These observations led to a detailed investigation of the role of one-dimensional glide of SIA clusters in controlling the damage accumulation under cascade damage conditions

[53–55]. The reasons as to why the concept of production bias is necessary for describing the damage accumulation under cascade damage conditions are discussed in [56].

It should be pointed out that the high rate of void swelling at low doses observed in copper (e.g. [57,58]) irradiated at 250°C could not be explained in terms of the SRT [59]. In fact, the SRT gave a swelling rate which was a factor of about twenty smaller than the experimental value. As mentioned above, this high swelling rate could be readily understood in terms of the PBM [41].

Over the years, a large number of investigations have demonstrated that under cascade damage conditions the vacancy accumulation in a zone along grain boundaries (henceforth referred to as "peak zone") is significantly enhanced (see [5] for a review), i.e. compared to that in the grain interior. The enhancement in the vacancy accumulation in the peak zone extends over distances of ~10–15 μm (~20 void spacings) from the boundary. The maximum in the enhancement seems to occur at about 10 cavity spacings from the boundary. Calculations of the swelling rate as a function of distance from a grain boundary demonstrated that the enhanced accumulation of vacancies in the peak zone cannot be explained in terms of the SRT and dislocation bias [60]. Recently, it has been shown that this behaviour can be understood in terms of the PBM and one-dimensional glide of SIA clusters produced in the cascades [54,55,61].

In this context it is interesting to note that the effect of grain size on void swelling during 1 MeV electron irradiation [62] has been found to be just opposite to that observed under neutron irradiation [63]. In the electron case, the swelling decreased (below the swelling value of the large grain) with decreasing grain size [63], whereas in the neutron irradiation the swelling even in the small grains was found to be considerably higher than in the large grains [63]. The results of 1 MeV electron irradiation were found to be fully consistent with the calculations in terms of the SRT model and dislocations bias [64]. It has been shown recently that the results of neutron irradiation, on the other hand, can be fully accounted for in terms of the PBM and one-dimensional glide of SIA clusters [65]. In fact, the analytical calculations show that the void swelling is proportional to $(R_g)^{-3/5}$, where R_g is the grain radius [65]. This means that under cascade damage conditions the effect of grain size on void swelling will be felt even in the large grains. Recent irradiation experiments on mono- and polycrystals (with different sizes) of pure copper with fission neutrons at 350°C have shown that indeed the swelling increases with decreasing grain size [65]. The results of numerical calculations using PBM and one-dimensional glide are in very good agreement with the experimental results.

In the earlier calculations [41,53–55] of damage accumulation in terms of the PBM and one-dimensional glide, a "mean size approximation" was used for the evolving clusters and cavities. This meant that the effects of the continuous transformation of sessile SIA clusters to the glissile ones (due to the prevailing vacancy supersaturation) on the details of damage accumulation could not be taken into account. Recently, this limitation has been overcome by using "size distribution functions" in the calculations of the evolution of sink strength, vacancy supersaturation and void swelling [66]. The calculated dose dependence of void swelling in terms of the PBM and one-dimensional glide of SIA clusters using size distribution functions for neutron irradiated copper at 250°C agrees very well with the experimentally measured results [66].

Since neither the intracascade clustering of SIAs nor the one-dimensional transport of SIA clusters can be treated within the framework of the SRT and BEK type models, the sensitivity of the damage accumulation to changes in recoil energies cannot be determined using these models. In contrast, one of the main predictions of the PBM is that the damage accumulation at a given temperature and damage rate should be sensitive to the efficiency of intracascade clustering of SIAs and vacancies and hence to the recoil energy. The validity of this prediction has been tested out experimentally by irradiating pure copper by 2.5 MeV electrons, 3 MeV protons and fission neutrons at 250°C. All irradiations were carried out at a damage rate of about 5×10^{-8} dpa (NRT)/s and up to a dose level of ~0.01 dpa (NRT). The dose dependence of the damage accumulation was determined using transmission electron microscopy and positron annihilation spectroscopy [47]. At the dose level of ~0.01 dpa, the electron irradiated specimens showed a complete absence of clusters. The cluster density in the proton and neutron irradiated (~0.01 dpa) specimens were found to be $\sim 7 \times 10^{20}$ and $3 \times 10^{22}/m^2$, respectively. The void density (at 0.01 dpa) also increased significantly with increasing recoil energy from $\sim 5 \times 10^{18}$ m^{-3} in the case of electron to $\sim 1 \times 10^{21}$ m^{-3} in the case of neutron irradiation.

The dose dependence of the void swelling results for electron, proton and neutron irradiations is shown in Fig. 7. These results clearly demonstrate that the damage accumulation is very sensitive to recoil energy (at least in the range used in the present experiments). An important implication of these observations is that the details of the damage production (i.e. intracascade clustering and surviving defect fractions, Fig. 6) play a very significant role in determining the level of damage accumulation, at least at low doses. To understand this effect of recoil energy, a detailed calculation has been carried out to determine the dose dependence of sink strengths, void size distributions, vacancy supersatura-

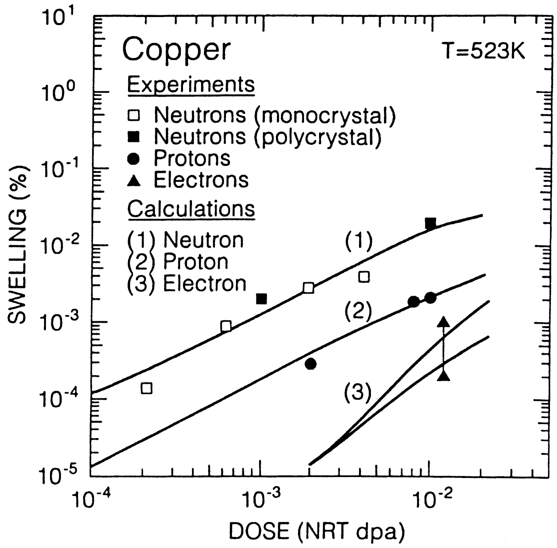

Fig. 7. Experimental and theoretical results illustrating the effect of recoil energy on void swelling in pure copper irradiated at 250°C with 2.5 MeV electrons, 3 MeV protons and fission neutrons [46,47]. The void swelling for the proton and neutron irradiations is calculated in terms of PBM using one-dimensional glide of SIA clusters, transformation of immobile (sessile) SIA clusters to glissile ones and size distribution functions. The void swelling for the electron irradiation is calculated in terms of SRT and dislocation bias. In both cases, the calculated results are in very good agreement with the experimental results.

tion and void swelling using the PBM, one-dimensional glide and size distribution function [46]. The calculated results agree very well with the experimental results. The calculated dose dependence of the void swelling is shown in Fig. 7. For comparison, the experimental results are also quoted in Fig. 7. It should be pointed out that the results for the electron irradiation are obtained using SRT and the present calculations yield a dislocation bias of ~2%.

These results demonstrate once again that the damage accumulation under Frenkel pair production can be easily understood in terms of SRT and dislocation bias. However, to understand the damage accumulation at higher recoil energies it is necessary to use the concept of production bias and one-dimensional transport of SIA clusters.

It is important to recognize that even though the calculations of damage accumulation carried out at present within the framework of the PBM include all the features of damage production outlined in Fig. 6, and can explain successfully a large number of experimental observations that could not be explained earlier in terms of the SRT and BEK type models, there still remain a number of problems that need to be solved before the model can become reliably predictive. For instance, serious efforts need to be addressed towards the following: (a) obtaining more accurate values for reaction cross-sections for gliding SIA clusters/loops with the sessile cluster/loops, grown-in dislocations, stacking fault tetrahedral (SFTs) and cavities, (b) determining the energetics of "absorption" of glissile loops into sessile loops and into grown-in dislocations and (c) solving the "old" problem of void nucleation.

As regards the damage accumulation under cascade damage conditions at relatively low temperatures (i.e. below the recovery stage V), very little seems to have been done. In fact, the work of Wiedersich [67] appears to be the only serious attempt addressed to this problem. Interestingly enough, Wiedersich's treatment [67] also recognizes the concept of production bias in that in his formulation he not only considers intracascade clustering of SIAs and vacancies but also makes an allowance for the fact that "the fraction of SIAs that form clusters in cascades is smaller than the corresponding fraction for vacancies". His calculations, however, do not include one-dimensional glide of SIA clusters.

While addressing the problem of irradiation hardening, Stoller has attempted to calculate the build-up of cluster density [68]. However, in this treatment the clusters of only di-, tri- and tetra-interstitials are considered. Furthermore, while calculating the build-up of cluster density, the fraction of interstitials contained in the form of clusters is set to zero. The vacancy clusters are assumed to be micro-voids instead of vacancy loops or tetrahedra.

It seems quite obvious, therefore, that a serious effort is required to address the problem of damage accumulation at low temperatures where the complications due to the presence of vacancy supersaturation and cavities do not exist. Recently, the question has been addressed in terms of kinetic Monte Carlo (KMC) simulations [69]. The computer simulations are performed using the stochastic annealing code ALSOME which is described in detail in [42]. The defect accumulation occurring during a continuous irradiation experiment is simulated [69] by successive introduction of defects and defect clusters (surviving in an isolated cascade at the end of the cooling-down phase of the cascade) in the simulation volume randomly in time and space. Cascade energies and the rate of their occurrence were chosen to mimic the damage conditions during irradiations with 14 MeV neutrons in the RTNS-II (rotating target neutron source) facility. The cascades were chosen from a library of cascades generated in MD simulations for recoil energies from 5 to 25 keV. Simulations included the effects of one-dimensional glide of SIA clusters containing 4–10 SIAs atoms.

The build-up of cluster density during irradiations with 14 MeV neutrons at room temperature was simulated as a function of dose up to ~0.1 dpa. The calculated cluster densities are found to be in a good agreement with the experimental results (see Fig. 1 in Ref. [69]).

4. Damage accumulation and materials performance

As indicated already in Section 1, experimentally it is well documented that neutron irradiation of metals and alloys at temperatures below the recovery stage V causes a substantial amount of hardening and a drastic decrease in ductility (i.e. low temperature embrittlement). Traditionally, the increase in the yield stress is considered to be caused by the defect clusters and loops accumulated during irradiation since the clusters and loops are assumed to act as barriers to dislocation motion. Originally, it was Seeger [70] who proposed that the "vacancy-rich zone" produced in the centre of a cascade may act as a barrier to gliding dislocations in the slip plane. Based on these considerations, a model, commonly known as dispersed barrier hardening (DBH) model was developed (see [71] for review). The model predicted that the increase in the yield stress due to irradiation should be proportional to the square root of the neutron fluence or of the product of the density and size of the loops accumulated during irradiation. It is rather surprising, however, that the problem of irradiation-induced loss of ductility (or embrittlement), which has been a matter of serious concern from the point of view of materials performance and lifetime in service, has not been treated theoretically.

In view of the available experimental evidence on the deformation behaviour and pre- and post-irradiation deformed and undeformed microstructures, the problem of irradiation hardening has been recently reanalyzed [72]. The analysis revealed that there are serious problems regarding the validity and applicability of the DBH model. In order to understand the processes involved in the initiation of plastic deformation in irradiated materials, a new model called Cascade Induced Source Hardening (CISH) has been proposed [72,73]. The main thesis of the CISH model is that during irradiation most of Frank–Read (F–R) sources (i.e. grown-in dislocations) get hardened by an atmosphere of small interstitial loops [73,74]. It should be mentioned that the possibility of the hardening of dislocation sources by "defect clouds" was considered by Blewitt et al. already in 1960 [75]. Because of the lack of detailed information regarding the damage production in cascades and the post-irradiation microstructure, they were not able to specify as to why and how F–R sources may be blocked. Detailed calculations [73,74] have shown, however, that the decoration of grown-in dislocations by small loops is likely to occur by trapping of glissile loops (produced in the cascades) in the strain field of the grown-in dislocations. A number of examples of dislocation decoration observed experimentally are shown in [73].

In order to initiate plastic deformation in the irradiated materials containing dislocations decorated with a cloud of small loops, these dislocations must be unlocked so that they can act as F–R sources. Hence, the stress necessary to unlock these dislocations must represent the upper yield stress. The break-away stress calculated using "small loop approximation" shows that the increase in the yield stress of neutron irradiated copper can be understood in terms of the CISH model [72–74]. Within the framework of this model, the main parameters controlling the upper yield stress are (a) the stand-off distance, y, between the edge dislocation and the row of sessile loops (decorating the dislocation), (b) the spacing, l, between the loops in the row and (c) the diameter, d, of loops in the row and the stress necessary to unlock the dislocations from the decoration is proportional to $(1/l)\,(d/y)^2$.

Even though at lower temperatures, the stand-off distance is not expected to change much with the irradiation dose, the dose dependence of the upper yield stress is complicated because both l and d will change with dose. The loop spacing, l, will decrease due to continuous loop accumulation, whereas the loop diameter, d, will increase due to loop agglomeration with increasing dose. These changes in the value of l and d is likely to depend on the flux of glissile loops to the decoration region. The flux will decrease with increasing dose as a result of the build up of clusters of SIAs and vacancies in the medium (i.e. between the decoration regions) and will reach a minimum when the cluster density in the medium will saturate. This would suggest that at least qualitatively, the upper yield stress would increase with increasing dose and would come to saturate at a certain dose level. Work is in progress to quantify these aspects of the dose dependence.

It is interesting to note that for the first time it seems possible to understand the irradiation-induced hardening as well as the decrease in ductility within the framework of one model, i.e. the CISH model. The decoration of the grown-in dislocations by the gliding SIA clusters produced in the cascades appears to be responsible not only for causing an increase in the upper yield stress, but also for initiating localized deformation in the form of "cleared channels". Experimental evidence on the dose dependence of the upper yield stress, frequency of cleared channel formation and occurrence of intergranular type of brittle fracture (see e.g. [36,76,77]) tend to suggest that the increase in the upper yield stress as well as the decrease in the ductility may be caused by the decoration of grown-in dislocations.

In view of the available experimental evidence it seems reasonable to suggest that the decoration of dislocations with SIA loops becomes more effective with increasing dose, causing a substantial increase in the upper yield stress. Under these conditions, dislocations can be generated only at the points of singularities with a high stress concentration factor. Once these sources are activated, they lead to the formation of cleared channels and most of the plastic deformation occurs in a very localized fashion in these channels. Meeting of these channels at grain boundaries, surfaces or other channels may lead to crack nucleation at these sites. Once the cracks are initiated, they are likely to propagate rapidly through the material causing intergranular fracture. Recently, the formation of cracks at grain boundaries has been demonstrated in 600 MeV proton irradiated pure iron [78].

Recently, low cycle fatigue experiments have been carried out at room temperature on pure OFHC-copper specimens irradiated with fission neutrons at ~50°C to a dose level of ~0.5 dpa (NRT) [79]. The cyclic stress–strain curve exhibits an upper yield stress followed by a yield drop. This is very similar to the behaviour observed during tensile testing of OFHC-Cu irradiated under similar conditions. The post-deformation microstructure shows the formation of cleared channels and the lack of dislocation generation in a homogeneous fashion during the low cycle fatigue experiments. These deformation characteristics would suggest that the grown-in dislocations get decorated with small SIA loops and are unable to act as dislocation sources even during the cyclic loading.

Thus, it is clear that the deformation behaviour of metals and alloys at lower temperatures (i.e. below stage V) is likely to be sensitive to the formation of glissile clusters. In the absence of glissile clusters/loops, the grown-in dislocations may not get decorated and deformation may proceed in a homogeneous fashion (i.e. without the formation of "cleared channels"). In other words, even the deformation behaviour may be sensitive to recoil energy and intracascade clustering of SIAs.

5. Summary and conclusions

In the preceding sections, various aspects of the production of atomic displacements and surviving defect fractions during irradiation with energetic particles have been considered. In particular, the influence of recoil energy on defect production efficiency, defect clustering in multidisplacement cascades and various fractions of surviving defects have been described. The significance of the complicated nature of the primary damage state containing mobile and immobile defect clusters of different thermal stability to the kinetics of the damage accumulation processes have been discussed. Finally, an attempt has been made to identify the role of damage production and accumulation in determining the mechanical performance of the irradiated materials. These considerations lead to the following conclusions.

- Undoubtedly, very significant progress has been made in the understanding of the primary damage state using MD simulations. However, some fundamental questions regarding the collapse of vacancy clusters into loops or stacking fault tetrahedra still remain to be answered. More dedicated efforts need to be made to establish the effects of alloying and strongly interacting impurity atoms on the evolution of the primary damage state.
- The problem of damage accumulation still remains a theoretical challenge. It is rather surprising that there exists practically no theoretical description of damage accumulation under cascade damage conditions at temperatures below stage V which includes considerations of gliding SIA clusters. For temperatures above stage V, the production bias model including one-dimensional glide of SIA clusters has been shown to be capable of describing the damage accumulation in pure metals. However, this model needs to be extended to cover low temperature and practical materials.
- The preliminary results of the kinetic Monte Carlo type of simulations of damage accumulation, using the primary damage state simulated by MD as input parameter, look quite promising. This technique can be used to simulate the damage accumulation at the low as well as high temperatures and can provide very useful support to analytical calculations of damage accumulation on a global scale.
- The present understanding of the irradiation-induced hardening and embrittlement at temperatures below stage V under cascade damage conditions remains tentative and qualitative. A considerable amount of work is necessary to establish a proper understanding of the commonly observed phenomena such as yield drop, lack of work hardening, plastic instability, and loss of ductility. Recently proposed explanation in terms of the cascade-induced source hardening (CISH) is still in a qualitative stage and a considerable amount of effort will be required to quantify the model.
- The considerations presented in this paper clearly suggest that the details of the damage production are important in determining the microstructural evolution as well as mechanical properties. It is crucial, therefore, that theoretical treatments aimed at describing the damage accumulation in the form of swelling and mechanical properties must take into account the effects of various primary damage production parameters.

- Finally, it must be emphasized that the basic and dedicated studies of damage production and accumulation are important not only for satisfying academic curiosities but are necessary for the longer term materials development programme for fusion reactor technology.

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