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PRODUCTION OF FREELY-MIGRATING DEFECTS DURING IRRADIATION*

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ABSTRACT

During irradiation at elevated temperatures, vacancy and interstitial defects that escape from their parent cascade and become free to migrate long distances can produce several different types of microstructural changes, e.g. void swelling, irradiation creep, radiation-induced segregation and precipitation, as well as the dissolution of existing metastable phases and concentration gradients. Hence the production rate of freely-migrating defects must be known as a function of irradiating particle species and energy before quantitative correlations can be made between microstructural changes occurring in different irradiation environments. Our fundamental knowledge of freely-migrating defect production has increased substantially in recent years. Critical experimental findings that led to the improved understanding are reviewed in this paper; areas that require further investigation are also identified.

A strong similarity is found for the dependence of freely-migrating defect production on primary recoil energy as measured in a variety of metals and alloys by different authors. The efficiency for producing freely-migrating defects decreases much more strongly with increasing primary recoil energy than does the efficiency for creating stable defects at liquid helium temperatures. The stronger decrease can be understood in terms of additional intracascade recombination that results from the nonrandom distribution of defects existing in the primary damage state for high primary recoil energies. Although the existing data base is limited to face-centered-cubic materials, the strong similarity in the reported investigations suggests that the same dependence of freely-migrating defect production on primary recoil energy may be characteristic of a wide variety of other alloy systems as well.

INTRODUCTION

Vacancy and interstitial defects that escape from their parent cascade and become free to migrate long distances can produce several different types of microstructural changes during irradiation at elevated temperatures, e.g. void swelling, irradiation creep, the dissolution of existing metastable phases and concentration gradients, as well as radiation-induced segregation and precipitation [1-4]. Hence the production rate of freely-migrating defects must be known as a function of irradiating particle species and energy before quantitative correlations can be made between microstructural changes induced at elevated temperatures in different irradiation environments. Such quantitative correlations will also contribute to our understanding of near-surface, materials property changes produced by ion-beam modification techniques.

Several sophisticated computer codes are available for calculating the total number of defects produced in various materials by different irradiation

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particles, i.e. electrons, ions and neutrons [5-8]. This is accomplished by first calculating the amount of energy transferred from the incoming particles to individual target atoms, then employing a damage function such as the Kinchin-Pease expression [9,10] to calculate the number of Frenkel pairs that are subsequently generated by recoiling target atoms of appropriate energies. Such dpa (displacements per atom) calculations have been used for many years as a first approximation to normalize microstructural changes produced in different irradiation environments [11,12].

However, as primary displacement events increase in energy from tens to hundreds of electron volts, defect production changes from the introduction of randomly spaced Frenkel pairs to the generation of several defects in close proximity to each other. Cascade regions of quite high defect density can be clearly identified in computer-simulation studies for primary-recoil energies as low as 1 keV [13,14]. Because the probability for subcascade formation increases with increasing primary recoil energy, the rate of increase in the spatial correlation among the irradiation-induced defects slows for recoil energies greater than several keV [15-17]. It has been recognized for many years that the higher defect densities generated at higher recoil energies will reduce the probability for defects to escape and become freely migrating. Hence the use of dpa calculations to normalize microstructural changes produced by different irradiation particles is obviously only a first-order approximation that cannot be expected to consistently yield quantitative agreement.

An improvement upon dpa calculations that takes into account the reduction in defect survival that occurs for higher recoil energies has been available for some years now, and is based upon measurements of defect production during irradiation at very low temperatures [18,19]. Low temperatures are employed for these experiments in order to maximize the number of irradiation-induced defects that are retained in the specimen. These measurements, which are discussed again later in the paper, demonstrate that the stimulated recombination of nearby vacancies and interstitials during the cascade cooling phase substantially reduces the probability for an irradiation-induced defect to survive an energetic cascade event. The low temperature findings indicate that dpa rates calculated for heavy-ion or neutron irradiations should be multiplied by a factor of approximately one-third for comparison with electron or low-energy, light-ion bombardment.

Several different experiments in recent years have shown that irradiations which generate energetically dense cascades are in fact much less than one-third as efficient as light-ion or electron irradiations at producing freely-migrating defects at elevated temperatures. Measurements at elevated temperatures of dislocation-pinning [20-22], anelastic relaxation [23], mass transport [24,25], ordering rates [26,27], and radiation-induced segregation [28-32] have been used to obtain information on the fraction of freely-migrating defects produced by a wide variety of irradiation particles. These measurements yield a dependence of freely-migrating defect production on primary recoil energy that is qualitatively similar to measurements of defect survival at low temperatures. However, the total fraction of defects that has been observed to undergo free migration at elevated temperatures for higher primary recoil energies is considerably less than the fraction of defects found to survive at low temperatures. This large difference has important implications for efforts to model or simulate microstructural changes induced in different irradiation environments.

In this paper, our current understanding of freely-migrating defect production is reviewed. Extensive work in our own laboratory using quantitative measurements of radiation-induced segregation to determine freely-migrating defect production at elevated temperatures is summarized. Pertinent findings reported by several other research groups are reviewed. A strong similarity is found for the dependence of freely-migrating defect production on primary recoil spectra as measured in a variety of metals and alloys by several different techniques. These results therefore provide a quantitative basis for correlating defect-flux driven microstructural changes produced in irradiation environments characterized by widely different primary recoil spectra. The similarity found for freely-migrating defect production in pure metals and different alloys suggests that the elevated temperature efficiencies can be applied with some confidence to yet uninvestigated alloy systems as well.

We begin in section II with a discussion of the primary recoil spectra for different types of irradiations and how these spectra are calculated. Low temperature measurements of defect production to determine dpa values are addressed in section III. Experimental determinations of freely-migrating defect production in our own and other laboratories are discussed in section IV. Conclusions are presented in section V, where recommendations for future work can also be found.

PRIMARY RECOIL SPECTRA

Several authors, on the basis of both experimental evidence and theoretical considerations, have noted that microstructural changes produced in different irradiation environments cannot be adequately normalized solely on the basis of dpa calculations. The underlying reason for the failure of the dpa normalization is quite simple. Displacement calculations determine the total amount of energy which goes into displacing atoms, but they ignore differences in the spatial distribution of the defect production. Since the number of jumps a defect makes before annihilation, i.e., the amount of atom transport or diffusion that occurs, depends strongly on whether the defects are produced in relatively isolated events or in cascade regions of locally high defect densities, a more detailed description that takes into account differences in the spatial arrangement of defects produced during irradiation is required to obtain a correct normalization. A partial description of this spatial arrangement can be inferred from the calculated distribution of energies with which the primary knock-on atoms recoil. This distribution is called the primary-recoil spectrum for an irradiation, and it can be calculated using several standard computer codes.

Since we are in fact concerned with describing the spatial distribution of the defects that are generated, the function of fundamental interest is the primary-recoil spectrum weighted by the total number of Frenkel defects subsequently produced by each primary recoil. This weighted recoil spectrum, which is discussed in more detail in Ref. 18, is obtained by calculating the fraction of defects, W(P) produced by all primary recoil-atoms with energies less than P. W(P) is given by

$$W(P) = \frac{1}{\nu_t(E)} \int_{P_{min}}^{P} dP' \, \frac{d\sigma(E,P')}{dP'} \, \nu(P'), \qquad (1)$$

where

$$\nu_t(E) = \int_{P_{min}}^{P_{max}} dP' \, \frac{d\sigma(E,P')}{dP'} \, \nu(P'); \qquad (2)$$

$d\sigma(E,P')/dP'$ is the differential cross section for a particle of energy E to produce a primary-recoil atom with energy P', and $\nu(P')$ is the number of Frenkel pairs generated by recoiling lattice atoms of energy P'. The integral in equation 1 is evaluated from the average threshold energy for atomic displacements, P_{min}, to energy P. The integral for $\nu_t(E)$, which yields the total cross section for Frenkel-pair production, is evaluated from P_{min} to P_{max}, the maximum energy transferable from a bombarding particle to a host atom.

W(P) as a function of P is shown in figure 1 for the irradiation of Cu by several different irradiation particles (1 MeV electrons, 200 keV and 1 MeV protons, and 2 MeV He, Ne, Ar and Kr ions); note that the abscissa is plotted as a logarithmic scale. The computer code PINTO [18], written by R. Benedek, was employed for the calculations, and an average threshold displacement energy of 29 eV was used. Frenkel-pair production was calculated using the modified Kinchin-Pease expression [10],

$$\nu(P) = \begin{cases} 0, & P < P_{min} \\ 1, & P_{min} < P < 2.5\,P_{min} \\ \dfrac{0.8E_D(P)}{2P_{min}}, & P > 2.5\,P_{min} \end{cases} \qquad (3)$$

where $E_D(P)$ is the damage energy. As the mass (or to a lesser extent, the energy) of the bombarding particle increases, the primary-recoil spectrum becomes harder. That is, more defects are produced in higher-energy recoil events, increasing the degree of spatial correlation among the defects.

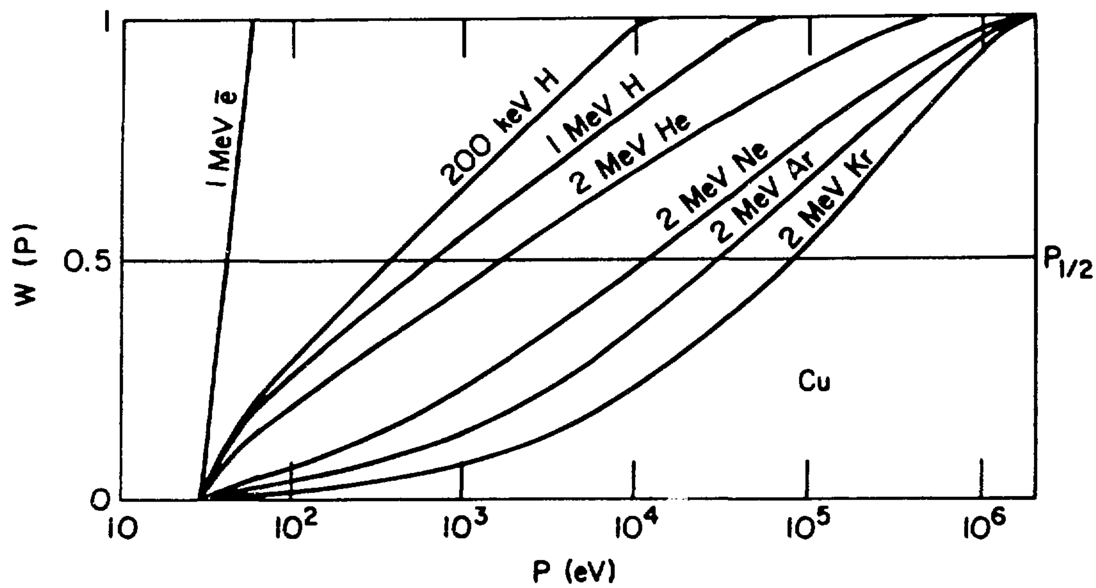

Fig. 1. Calculated fraction, W(P), of defects produced by primary recoil events of energy less than P for several different irradiation particles incident on copper.

The maximum energy that can be transferred to a copper lattice atom by 1 MeV electrons (figure 1) is slightly less than 60 eV. Since no defects are produced below the minimum threshold energy of 29 eV, the range of recoil energies during 1-MeV electron irradiation of Cu is very narrow (29-60 eV), and only isolated Frenkel pairs are generated. However, because ions can transfer large amounts of energy via direct collisions as well as small amounts via long-range Coulombic interactions, both relatively low-energy, light-ions and high-energy, heavy-ions produce a wide spectrum of primary recoil energies. The maximum energy that can be transferred is given by $(4m_1m_2)\ E\ (m_1+m_2)^{-2}$, where m_1 and m_2 are the masses of the ion and target atom, respectively, and E is the incident ion energy.

No single parameter describes defect production during irradiation as completely as the weighted primary-recoil spectrum does. However, the value of P where W(P) = 0.5 provides one measure of the physical "hardness" of a given recoil spectrum. This quantity, i.e., that primary-recoil energy above and below which half of the defects are produced, is referred to as the weighted-average recoil energy, $P_{1/2}$. The weighted-average recoil energy will be used later in this paper to quantify measurements of the efficiency for producing freely-migrating defects during irradiation with particles of different masses and energies.

$P_{1/2}$ values for the ion irradiations of copper listed in figure 1 vary from 380 eV for the 200 keV protons to more than 80 keV for 2 MeV Kr bombardment. An intuitively useful quantity, the number of Frenkel pairs produced during an "average" recoil event can be obtained by substituting values of $P_{1/2}$ into the modified Kinchin-Pease damage expression (equation 3). In this fashion, we see that by varying the ion energy over one order of magnitude (from 200 keV to 2 MeV) and by using ions with masses ranging from 1 to 84 amu, we can vary the number of defects produced in the average recoil event from as little as ~5, to greater than 1000. Since these changes in mass and energy can be accomplished with a single ion accelerator, ion irradiation constitutes a powerful and versatile experimental technique for investigating the effects of different primary recoil spectra on irradiation phenomena.

The influence of target mass on the weighted-average, primary recoil spectrum is addressed briefly in figure 2, where results calculated for 1 MeV protons and 2 MeV Kr ions incident on Al and on Au are displayed. Threshold displacement energies of 27 and 43 eV were employed for Al and Au, respectively. As could be anticipated from knowledge of the maximum energy that can be transferred, effects of target mass on $P_{1/2}$ become most pronounced when the mass of the incoming ion nears that of the target atoms. Also shown in figure 2 is a typical, weighted primary recoil spectrum for irradiation of nickel by reactor neutrons [33]. Since neutrons interact with matter only over very short distances, the value of $P_{1/2}$ is relatively large (50 keV), despite the small mass of the neutron. Hence neutron irradiation in general produces a much higher fraction of energetically dense cascades compared to ion irradiation.

The hardness of neutron relative to ion irradiations becomes even more apparent when it is realized that primary knock-on atoms with very high recoil energies do not necessarily generate correspondingly denser, individual cascades. Both experimental and theoretical studies have shown that the spatial correlation among the defects does not increase indefinitely with increasing primary-recoil energy. Transmission-electron-microscopy studies by Merkle et al. [15] first demonstrated that very energetic recoils create closely-spaced, but clearly resolvable, subcascade regions. Development of subcascades was postulated by Averback et al. [18] to explain their experimental observation of saturation in the low-temperature, irradiation-induced resistivity increment at very high primary-recoil energies. Theoretical calculations of displacement cascades by Jan [16] and by Sigmund [17], as well as computer-simulation studies by Robinson and Torrens [13] and by Beeler et al. [14] also indicate a high probability for subcascade formation at primary-recoil energies above a few tens of keV. Hence as recoil events increase in energy above many keV, the increase in spatial correlation among the defects with increasing recoil energy slows because of the increasing probability for subcascade formation.

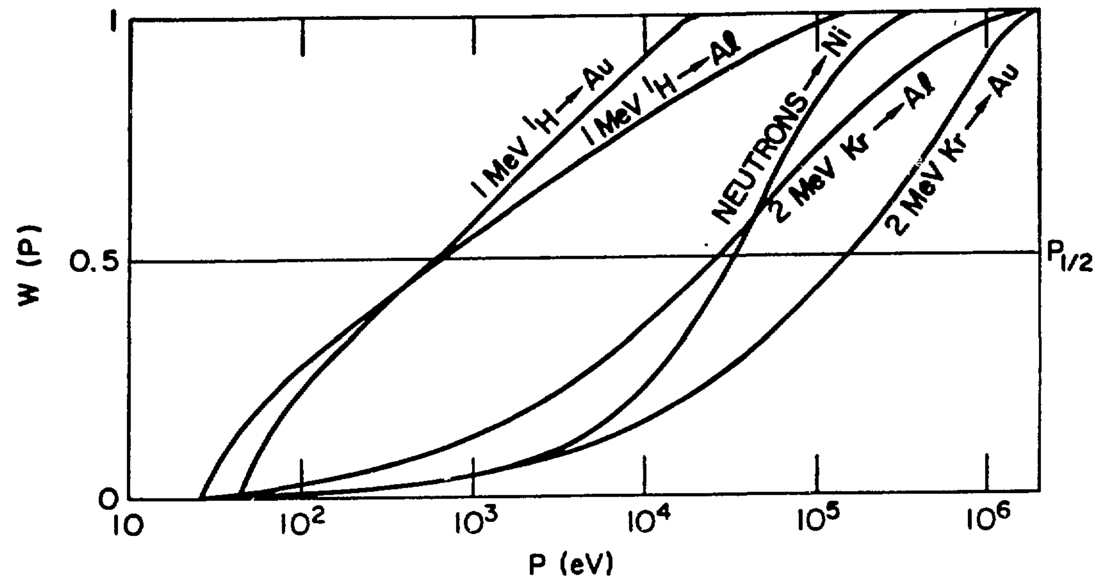

Fig. 2. Fraction of defects produced by primary recoil events of energy less than P for five different irradiations.

DEFECT PRODUCTION AND DPA

For many years, measurements of various property changes during irradiation at liquid helium temperatures have been performed to determine the number of Frenkel defects that are produced by different irradiation particles [34]. Because of their experimental simplicity and high sensitivity, electrical resistivity techniques are commonly used [35]. Low temperatures are maintained to minimize the migration of defects subsequent to production, and therefore to maximize the fraction of defects that survive. The number of stable Frenkel pairs is obtained by dividing the total irradiation-induced property change by that due to a single Frenkel pair, a quantity that must be determined by independent measurement or calculation. Studies at these low temperatures provide information on what is referred to as the primary damage state [36], i.e. the defect configurations that exist in the lattice after the short-term annealing that occurs during the cascade cooling phase is completed.

As commonly used, the term displacements per atom (dpa) refers to the number of times a target atom has received sufficient energy to become a stable interstitial defect, i.e. one that would remain indefinitely in the specimen at temperatures near absolute zero. Note that this definition of dpa, which is quite useful for describing certain aspects of defect production, is misleading in terms of net atom transport during irradiation.

Although they may not become part of a stable Frenkel pair in the process, atoms in fact do exchange lattice sites many times more often during irradiation than is indicated by the number of dpa. For example, molecular dynamics simulations reveal that many Frenkel pairs created within energetically dense cascades spontaneously recombine, i.e. without thermal activation, during the initial picosecond of a cascade event [37,38]. Those defects that spontaneously recombine can produce several exchanges of atoms between lattice sites, but these exchange events are not included as part of the calculated dpa number. Futhermore, replacement collision sequences can result in many atomic replacements for each permanently displaced interstitial. Disordering and ion-beam mixing experiments have shown that the number of atomic replacements per dpa in energetically dense cascades is on the order of 100 [39]. The fact that atom transport jumps can be as much as one or two orders of magnitude greater than the dpa number, depending on whether defects are produced in cascade events or as isolated Frenkel pairs, is yet another indication that attempts to correlate irradiation-induced microstructural changes on the basis of dpa values can not be expected to achieve quantitative agreement.

Experimental and theoretical investigations of defect production at low temperatures have been reviewed elsewhere, including at these proceedings [39]. We therefore discuss only a few general observations that will be pertinent to the discussion of freely-migrating defect production. Averback et al. [18] systematically investigated defect production during ion irradiation at 4 K in copper and silver over a wide range of primary recoil energies, from about 100 to almost 10^6 eV. Kinney, Guinan and Munir [19] performed a similar study in a number of pure metals irradiated with electrons, ions and neutrons. Both investigations found a strong decrease in the efficiency of defect production with increasing recoil energies up to values of a few kilo-electron-volts; this result clearly shows that cascade effects on defect production become important at relatively low recoil energies (1-2 keV). In both studies, the efficiency for producing stable defects at 4 K appeared to saturate at high recoil energies, at a value of about one-third that found for the lowest primary recoil energy. The observed tendency for saturation demonstrates that the energy density within cascades does not increase indefinitely with increasing primary recoil energy. The results of an extensive interlaboratory program to study low temperature damage rates in dilute alloys and pure metals ~~have~~ been summarized by Jung et al. [8]. Dilute alloying additions appear to produce no substantial effects during irradiation at these low temperatures, where thermally activated defect migration can be neglected.

PRODUCTION OF FREELY-MIGRATING DEFECTS

Freely-migrating defects lead to mass transport over distances large compared to cascade dimensions only at temperatures (typically > 600 K) where both vacancy and interstitial defects, and their small clusters, are relatively mobile [40]. Very little information exists for extrapolating the low-temperature results described in the previous section to such elevated temperatures. Experimental techniques with sufficient temporal ($\sim 10^{-11}$s) and spatial (~1-10 nm) resolution are not currently available for investigating the primary damage state (cf sec. II) at temperatures where one or more defect types are mobile. In fact, while a primary damage state for a cascade event can clearly be defined after low-dose, low-temperature irradiation, it is not clear that an analogous state even exists at high temperatures, where the defects are always mobile.

At elevated temperatures, additional recombination and clustering will occur within cascade regions, and some defects will escape and migrate freely through the lattice. The argument is often made that since defect production involves energies (20-100 eV) that are so much larger than thermal energies, little effect of temperature is expected on displacement processes. However, indications of substantial effects, for example on the threshold displacement energy [41], have been reported. In any case it should be emphasized that even in the absence of strong effects of temperature on the production process, the low-temperature results discussed in section III provide a basis only for determining the number of defects that are present in the specimen following cessation of the athermal part of the cascade event ($\lesssim 10^{-11}$s). Obviously the defects that result from cascade production are not, at this stage, randomly distributed throughout the sample, but in fact the positions of the same as well as dissimilar types of defects are strongly correlated. Hence, a more direct method is needed to determine the production efficiencies of freely-migrating defects during irradiation with different particles at elevated temperature.

In recent years, several authors have taken more direct experimental approaches. The strategy is quite simple; one of several mass transport effects that are driven by freely-migrating defects is characterized by careful, quantitative experimental measurements under a variety of steady state, or quasi-steady state, irradiation conditions. Then theoretical modeling and/or empirical relationships are used to extract the fraction of freely-migrating defects that contributes to the measured mass transport for different primary-recoil spectra. In this section we review these measurements, beginning with studies of radiation-induced segregation (RIS) in two concentrated alloys that were performed in our laboratory.

In the first study, Rutherford Backscattering Spectrometry (RBS) was used to investigate RIS in concentrated Ni-12.7 at.% Si alloys [28]. In agreement with simple theoretical arguments, the thickness of nonequilibrium coatings of Ni_3Si that were formed on the irradiated surfaces by RIS was found to increase linearly with the square root of the irradiation time at a constant dose rate, and to exhibit a temperature dependence characteristic of freely-migrating defects [42]. In the recombination-limited regime, the growth-rate of the coatings was found to vary inversely as the fourth root of the dose rate [43].

During irradiation with different ions at approximately the same calculated dpa rate, the growth rate of the coatings was found to decrease rapidly with increasing ion mass. For the range of experimental temperatures (650-900 K) that were utilized, effects of different irradiation-induced sink structures by different mass ions could be eliminated as a significant source for the decrease.

From accurate measurements of coating growth rates, and the empirically determined and theoretically understood dose-rate dependence, the relative efficiencies of the different ions for producing freely-migrating defects, i.e. those defects responsible for the RIS, could be extracted. The results [28] are shown in figure 3, where the relative efficiencies (normalized to that of 1 MeV protons) are plotted as a function of the defect-production-weighted, average recoil energy, $P_{1/2}$; the abscissa is plotted as a logarithmic scale.

Figure 3 provides a quantitative representation of the relative efficiency for producing long-range migrating defects in Ni-Si alloys (open circles) as a function of the "hardness" of the primary-recoil spectrum. The data represented by the filled circles will be discussed shortly. Irradiations with weighted-average recoil energies of 1.8, 2.7, 51, and 74 keV are, respectively, only 48%, 37%, 8% and <2% as effective as an irradiation with a weighted-average recoil energy of 730 eV for introducing defects which are free to migrate long distances. The most rapid decrease in efficiency occurs over recoil energies up to about 5 keV. Note that the general shape of the curve in figure 3 is qualitatively very similar to the low-temperature, defect survival results discussed in Section III. In particular, a strong decrease occurs in the efficiencies of both processes with increasing recoil energy up to values of a few keV. Furthermore, the efficiencies of both processes appear to saturate at high recoil energies.

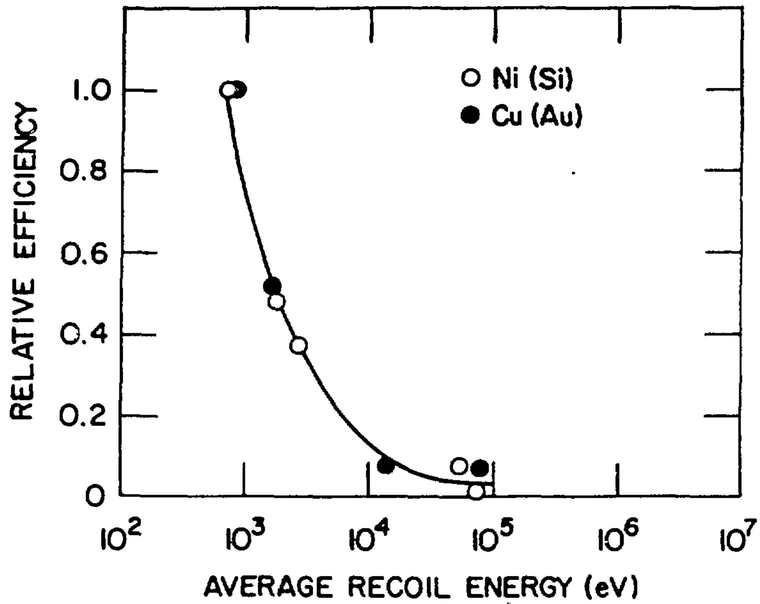

Fig. 3. Relative efficiencies of various ions for producing long-range, freely-migrating defects plotted as a function of the calculated, weighted-average recoil energy.

However, a large quantitative difference is readily apparent. The low-temperature, defect survival measurements saturated at an efficiency which was approximately 1/3 of that found at the lowest primary-recoil energy. In contrast, the apparent limiting value in figure 3 is substantially less, only a few percent of that found at the lowest primary-recoil energy. The observation that the efficiencies for producing freely-migrating defects decrease to considerably lower values than the defect production and survival measurements do, implies that the fraction of freely-migrating defects is also inversely related to the hardness of the primary-recoil spectrum. Defect interactions within cascades cause the fraction of freely-migrating defects to decrease even faster with increasing average recoil energy than does the total number of defects surviving in the primary damage state. This additional intracascade recombination at elevated temperature apparently occurs subsequent to the time ($\sim 10^{-11}$ s) required to achieve the primary damage state of a cascade at low temperatures. The strong decrease in efficiencies is in good agreement with computer simulation studies of intracascade annealing by Muroga et al. [44,45] over the entire range of recoil energies employed in their calculations (30 eV-30 keV).

Although the efficiency results are supported by additional evidence provided by several other investigators that is discussed below, the possibility existed that the very low efficiencies found at high recoil energies might result either from the known strong interaction between Si and interstitial defects in Ni-Si alloys, or to the second-phase formation that occurs [46]. For these reasons, we have recently performed similar measurements of RIS in a Cu-1 at.% Au alloy [47]. Au is an oversize solute in Cu, and the interaction between interstitial defects and Au atoms is relatively weak. As a result interstitial defects do not preferentially transport solute (Au) atoms toward defect sinks in Cu-Au alloys, as they are believed to transport Si atoms in Ni-Si alloys. In fact, Au depletion is observed at the irradiated surface, demonstrating preferential transport of solvent (Cu) atoms by the interstitial flux. Cu-Au alloys were also selected for the measurements because accurate RBS measurements required only about 1 at.% Au in the alloy, minimizing the possibility of second-phase formation.

The efficiencies determined for producing freely-migrating defects in Cu-Au are shown by the filled circles in figure 3, alongside the Ni-Si results. Despite the very different solute-interstitial interactions in the two alloys, and despite the formation of a second phase in one but not the other alloy during irradiation, the relative efficiencies for producing freely-migrating defects extracted from the Cu-Au and Ni-Si RIS measurements exhibit a practically identical functional dependence on primary recoil energy.

We now turn to a discussion of results on freely-migrating defect production reported by other authors. These additional measurements substantially extend the range of primary recoil energies over which information on freely-migrating defect production is available. Some of the earliest determinations involved reactor irradiation studies at relatively low temperatures. Schilling et al. [48] estimated from resistivity recovery performed just above stage I (~50 K) that > 15% of the interstitials produced by fast-neutron irradiation of Cu escape from the displacement cascades. This percentage is in excellent agreement with the results of Theis and Wollenberger [49], who used resistivity measurements to monitor defect production during reactor irradiations between 50 and 170 K, and report that ~15% of the number of observed interstitials in copper escape any correlated reactions. Since the total number of defects observed by resistivity techniques decreases by about a factor of 3 in going from low-recoil-energy bombardments to reactor irradiations, these latter two studies indicate that the reactor irradiations (typical average recoil energy of 30 keV) are only about 5% as efficient as low-recoil-energy irradiations at creating freely-migrating defects. Beretz et al. [23] have used strain relaxation measurements to study defect production in Ag-24 at.% Zr by reactor neutrons at temperatures between ~300 and 400 K. From their calculation of the total defect production, they conclude that only ~10% of the defects created in the displacement cascades escape annihilation and/or agglomeration and become freely migrating. A semiquantitative analysis by Kirk and Blewitt [26] of experimental results by Blewitt et al. [27] implies that fast neutrons (typical recoil energy of ~20 keV) are only a few percent as effective as thermal neutrons (typical recoil energy of ~300 eV) at producing migrating vacancies in Cu_3Au at 423 K. Goldstone et al. [21,22] have studied dislocation pinning in copper during electron and neutron irradiation at temperatures between 300 and 400 K. They find that fast neutrons in the energy range from 2 to 20 MeV are again only a few percent as effective as (0.5-1)-MeV electrons (average recoil energy ~50 eV) at producing interstitials which migrate to pin dislocations. All these measurements support the idea that the relative efficiency for creating long-range migrating defects drops to only a few percent of the initial value as the primary-recoil energy increases from several tens of electron volts to many kilo-electron-volts.

The low to intermediate irradiation temperatures employed in the examples cited directly above mean that the irradiation-induced, defect-sink structure may play an important role in determining the net amount of mass transport that is measured and subsequently used to determine the efficiency for producing freely-migrating defects. By going to still higher irradiation temperatures, where defect agglomerates are also unstable with respect to annealing, the complications from the irradiation-induced sink structure can be avoided, as was done for the RIS measurements discussed earlier. We will now discuss further measurements performed in this higher temperature range.

The Berlin group [24,25,50,51] has published several studies of radiation-enhanced self-diffusion in nickel, and of radiation-enhanced solute diffusion in Ni, Cu, and Fe-Cr-Ni alloys during ion irradiation in the temperature range from ~300 to 1000 K. The observed temperature and dose-rate dependencies of the steady state defect concentrations in the 600 to 1000 K range are within experimental error identical to those found in the RIS studies, demonstrating that effects from the irradiation-induced sink structure are negligible under these irradiation conditions. For 300 keV Ni ion irradiation, these authors conclude that the production rate of freely-migrating defects, i.e. those that contribute to the radiation-enhanced diffusion, is only ~2.5% of the calculated dpa rate. For 300 keV self-ion irradiation of Cu, they find a value near 5%, and ~4% for 4 MeV Ni ions. Similarly low values were deduced for freely-migrating defect production from rate-equation fits by Marwick et al. [30] to their experimental results on RIS in Fe-Ni-Cr alloys during 300 keV Ni ion irradiation at temperatures between 700 and 950 K. The weighted-average primary recoil energies for all these irradiations are $\gtrsim$ 50 keV.

Muroga et al. [52] employed stereo-electron-microscopy measurements of the thicknesses of radiation-induced, surface precipitate layers to determine the efficiency of 1-MeV electrons for producing freely-migrating defects in Ni-12.7 at.% Si. Their value shows that the efficiency approximately doubles in going from 1-MeV protons to 1-MeV electrons. All defects generated by 1-MeV electrons are created by primary recoils < 80 eV, i.e., the 1-MeV-electron irradiation generates predominantly single, isolated Frenkel pairs. However, many defects generated by the 1-MeV protons are produced in cascade events. For example, about 25% of the defects are produced in events with energies greater than 5 keV. It is these higher-energy-recoil events which presumably are the major factor in reducing the efficiency of 1-MeV protons to only one-half that of 1-MeV electrons.

The Curie temperature of Ni-Si alloys is a sensitive function of the Si concentration in solid solution. This effect has been employed to study RIS during MeV electron irradiation by Barbu et al. [31], and in an identically prepared specimen during 14 MeV neutron irradiation by Huang and Guinan [32]. The results of the two experiments are shown in figure 4, where the measured increases in the Curie temperature during irradiation, which occur as a result of the removal of silicon from solution by RIS to defect sinks, are plotted as a function of the calculated energy deposited in atomic displacements. Clearly the transport of Si atoms to defect sinks during neutron irradiation is much less efficient per number of displaced atoms than that during electron irradiation. Although the very small effect during neutron irradiation is difficult to quantify, the slope obtained from the author's fit to their data is about one-tenth that for the electron irradiation results. Assuming an inverse fourth-root dependence on dose rate of the amount of silicon removed from solution, the results in figure 4 imply that freely-migrating defect production during 14 MeV neutron irradiation is only 6-7% of

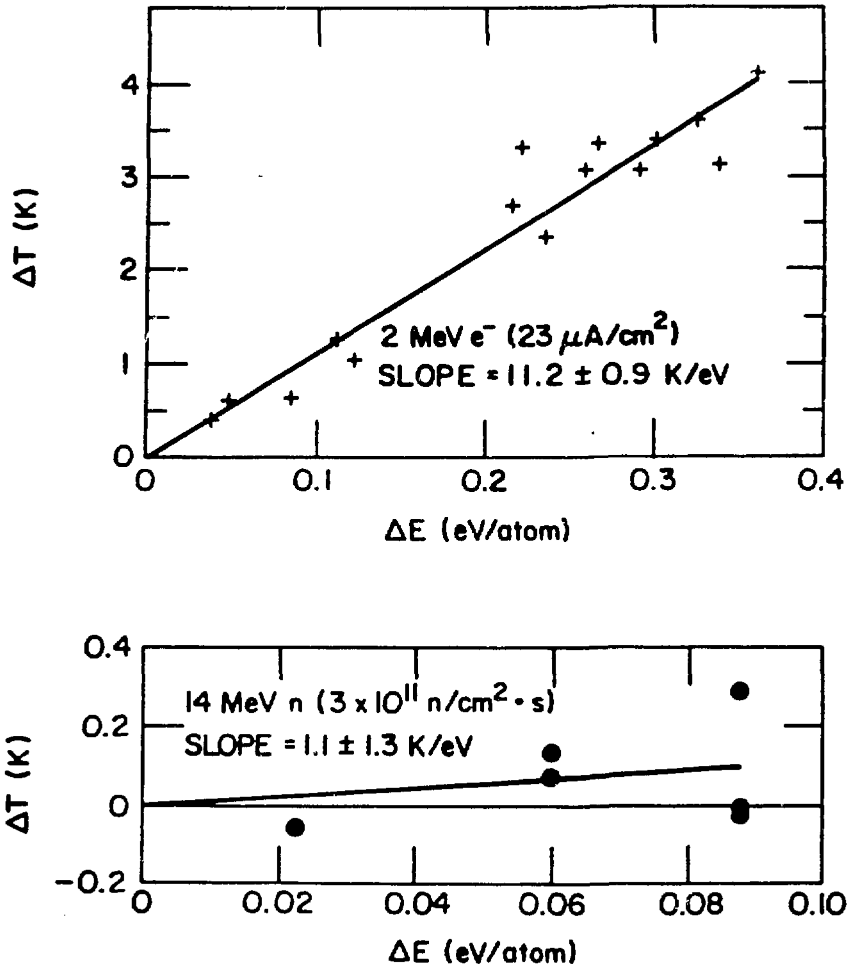

Fig. 4. Increase in Curie temperature of a Ni-4 at.% Si alloy measured during 2 MeV electron (top, Ref. 31) and 14 MeV neutron (bottom, Ref. 32) irradiation at 623 K. The calculated dose-rate during the electron bombardment was 4.5×10^{-7} eV/s, and 8.9×10^{-8} eV/s during the neutron irradiation.

that during 2-MeV electron bombardment at the same calculated dpa rate. The weighted average recoil energy is approximately 60 eV for the 2 MeV electrons, while that during 14 MeV neutron irradiation is 500 keV.

SUMMARY AND CONCLUSIONS

Microstructural changes induced by mass transport during irradiation at elevated temperatures are driven primarily by those defects that escape from their parent cascade and become free to migrate long distances. Such microstructural changes include irradiation creep, void swelling, radiation-enhanced diffusion phenomena, and radiation-induced segregation and phase redistribution. Several currently available measurements involving mass transport effects during irradiation at elevated temperatures provide definitive evidence that the efficiency for producing freely-migrating defects decreases strongly with increasing primary recoil energy. In particular, the net decrease is substantially greater than that found for the production of stable defects during irradiation at very low temperatures. This larger decrease has important implications for efforts to theoretically model or experimentally simulate microstructural changes induced at elevated temperatures in different irradiation environments.

Only a few percent of the total number of defects that exist in the primary damage state produced by energetic recoil atoms actually escape and undergo free migration. The stronger decrease in freely-migrating defect production is readily understandable in terms of the primary damage state introduced by energetic displacement cascades, which contains a highly nonrandom defect distribution. Because of this stronger reduction in the fraction of freely-migrating defects at higher recoil energies, dpa calculations that simply describe the number of stable defects generated during irradiation are inadequate for correlating microstructural changes that occur in different elevated temperature irradiation environments. However, the quantitative understanding of freely-migrating defect production as a function of primary recoil energy that has emerged in recent years does provide a physically realistic basis for deriving these correlations. In addition, the strong similarity observed in the functional dependence of freely-migrating defect production on primary recoil energy in different alloyed and unalloyed metals implies that this same functional dependence may also be characteristic of uninvestigated alloy systems as well.

However, a certain degree of caution must accompany the last observation. In fact, although similar dependencies have been noted in several materials, the investigated systems are limited at present to face-centered-cubic metals. Hence one obvious area for future investigation is the extension of freely-migrating defect production measurements to a wider variety of alloy systems. We are currently investigating body-centered-cubic and amorphous systems in our own laboratory, using in situ measurements of radiation-induced segregation effects.

Another quite obvious area for future work is the determination of absolute efficiencies. The RIS results discussed in this paper (figure 3) constitute the most systematic study to date of freely-migrating defect production at elevated temperatures. The results, however, are reported as relative values. Current attempts to obtain absolute values employ fits [23,24,30] of the experimental data to model calculations that require several poorly understood input parameters. Hence the accuracy of the absolute efficiencies that are currently available are highly uncertain.

A deconvolution of the relative efficiencies, which are expressed in terms of the "weighted-average" primary recoil energy, into a function of the primary knock-on atom recoil energy would also be highly desirable. This would allow a more accurate identification of when subcascade formation becomes an important factor in the generation of freely-migrating defects within displacement cascades. Similar deconvolutions of low-temperature, defect production measurements have been performed, for example by Jung et al. [8]. It should be admitted, however, that much less quantitative information is available on freely-migrating defect production at elevated temperatures than exists for defect survival probabilities at low temperatures, and therefore the completion of this task will presumably await the availability of more extensive experimental results. Neutron irradiation results, because of their characteristically narrow range of recoil energies, would be particularly useful.

Another area that deserves mention in terms of future work is the determination of the probabilities for vacancies to escape cascades relative to those for interstitials. Of course at sufficiently elevated temperatures, where none of the cascade agglomerates such as loops or clusters are stable, these probabilities must be equal. However at intermediate temperatures, where certain types of defects are mobile on the time scale of the experiment but others are not, the probabilities can in fact be significantly different. Since one type of defect may contribute proportionately more to the measured experimental change, say vacancies to reordering or interstitials to segregation phenomena, it is possible to determine these different probabilities in appropriately selected alloys. In fact, isolated examples of determinations of these quantities exist, but no systematic studies have been performed over a broad spectrum of recoil energies.

Finally, more accurate determinations are needed of mass transport processes during irradiations that produce a high proportion of energetically dense cascades. It is clear that because only a small percentage of such defects actually become freely-migrating, the effects of such irradiations on mass transport processes are small and therefore difficult to measure accurately. In addition, the small fraction of freely-migrating defects produced by energetic displacement cascades makes it difficult to obtain dose rates equivalent to those achieved during light ion bombardment, making it necessary also to determine the dose-rate dependence of the measured effect very accurately. However, a difference between two and four percent in the fraction of freely-migrating defects translates directly into a factor of two difference in the input parameter for modeling calculations, making accurate values very important. Again, well-conceived neutron irradiation experiments appear particularly attractive.

During the past few years, considerable progress has been made in our fundamental understanding of freely-migrating defect production during irradiation at elevated temperatures. This understanding is providing a basis for quantitative correlations of microstructural changes produced in quite different irradiation environments. Such correlations will greatly enhance the usefulness of ion and electron irradiation experiments, which have the advantage that they can be performed under highly flexible but tightly controlled experimental conditions, for predicting materials behavior in neutron irradiation environments. In addition, this knowledge will contribute to the optimization of near-surface materials properties produced by ion-beam modification techniques.

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