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Void nucleation with embryo injection

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Abstract

The effect of embryo injection on void nucleation is evaluated through an extension of classical nucleation theory. Calculations indicate that embryo injection may play a significant role in void nucleation under cascade-producing irradiation environments, such as occur in fission and proposed fusion reactors.

1. Introduction

Void swelling was discovered nearly 40 years ago in the course of materials development for the liquid metal fast breeder reactor cladding and duct materials [1,2]. Vast numbers of nanometer to micrometer diameter, roughly equiaxed voids formed, giving rise to volumetric swellings of tens of percent. First wall materials in proposed tokamak-type thermonuclear reactors are to be exposed to similar irradiation conditions, hence are likely to experience void swelling. In addition it has recently been found that some internals in light water reactors undergo void swelling after long exposure times [3]. Void swelling is thus a major consideration in most kinds of nuclear power reactors.

Cascades are neither a necessary nor a sufficient condition for void formation. Electron irradiation readily produces displacement damage, but in the form of single Frenkel pairs, so that no cascades are produced. Nonetheless, electron irradiation has been found to produce void swelling in a wide variety of alloys. On the other hand, gold and titanium were found not to swell, even under cascade-inducing irradiation [4].

The irradiation environment in nuclear materials is so complex that even today no rigorous, complete theory of void nucleation exists. This paper expands the theory for void nucleation from a sea of vacancies and self-interstitials to include the effects of displacement cascade-induced vacancy clusters as void embryos.

2. The irradiated state

Neutron energies in fission and fusion reactors typically range from the so called fast (MeV) range all the way down to thermal neutrons with energies

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the order of kT (<0.1 eV). The lower energy neutrons usually play a secondary role in atomic displacements but may give rise to atomic transmutations, in particular (n,α) reactions.

Newtonian mechanics show that an energetic neutron my transfer at most about 7% of its energy to (for example) an iron atom. If the struck atom receives enough energy to displace it from its lattice site it is known as a PKA, or primary knock on atom, which proceeds through the lattice, transferring energy to other atoms. Ultimately the PKA will come to rest leaving behind it a trail of collision partners. Some of these struck atoms may also leave their sites, becoming secondary knock on atoms, which in turn transfer energy to other lattice atoms.

Investigators in the 1950s and 1960s [5–7] depicted the PKA as giving a shower of vacancies and self-interstitials, with a central, highly disordered region sometimes referred to as a spike. The entire event was called a displacement cascade, which was often thought to contain a core of clustered vacancies. It was also realized that in some cases the struck atoms would not entirely leave the lattice site, or would be displaced to a nearby site from which recombination could occur very quickly.

Analyses of radiation effects in the 1970s usually assumed that the displacement cascade quickly dissipated into a uniform sea of single vacancies and self-interstitials [8,9]. Most workers assumed that any vacancy or interstitial clusters formed quickly evaporated into single defects. Others, as far back as Levy [10] suggested that vacancy clusters might provide precursors for void nucleation. These suggestions were anecdotal: the present work is the first attempt to put the role of cluster injection on void nucleation on a quantitative basis.

Some 20 years ago Seidman and co-workers (see Wei et al. [11]) studied field ion microscope tips that had been irradiated with energetic heavy ions, which performed the role of the PKA. The irradiations were at such low temperatures (<10 K) that the self-interstitials and vacancies were both immobile. These workers were able to locate the position of every vacancy surviving cascade relaxation. They did not concern themselves with the locations of the self-interstitials. Fig. 1 shows the results for tungsten irradiation with a 20 keV W^+ ion.

The figure shows that many of the vacancies have one or more other vacancies as nearest neighbors. It is clear that given enough thermal energy for vacancy movement, some of the vacancies would cluster into compact aggregates that might serve as precursors in void nucleation.

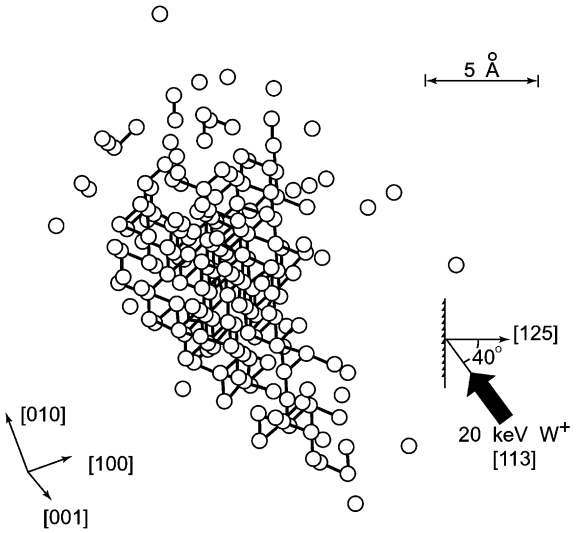

Fig. 1. Vacancy-rich zone in W caused by a 20 keV W^+ ion incident at the angle shown. Circles connected by lines indicate vacancies on nearest neighbor sites. (Figure courtesy of Dr D. Seidman.)

The key ingredients of molecular dynamics (MD) simulations: hardware, software, and interatomic potential energy functions have undergone huge advances in the last decade or so. Even so the computer demands of MD are so huge that even on the most powerful machines the simulations describe at most ~100 ps of real time for a region of $\sim 10^6$ atoms. Fortunately this combination of number of atoms and time is enough to give a very accurate description of displacement cascade formation and the very earliest stages of annealing [12–15].

Fig. 2 shows the development of a cascade in Fe from a 2 keV PKA. Fig. 2(a) shows an early stage in cascade development at 0.075 ps, followed by the maximum level of displacement at 0.34 ps. Spontaneous recombination then occurs, as shown by views at 0.59 ps and 9.8 ps. The final view is at the order of 100 lattice vibration times, so that very little thermally activated motion has taken place. The order of 500 atoms are displaced at the maximum, at 0.34 ps, but only 28 displacements survive at 9.8 ps.

Fig. 3 illustrates a 20 keV cascade in Fe after 20 ps and then at 200 ps, after the very early recombinations have taken place. It was found that the order of one fourth of the self-interstitials created were in the form of clusters ranging in size from di-interstitials up to dozens of interstitials. The binding energy

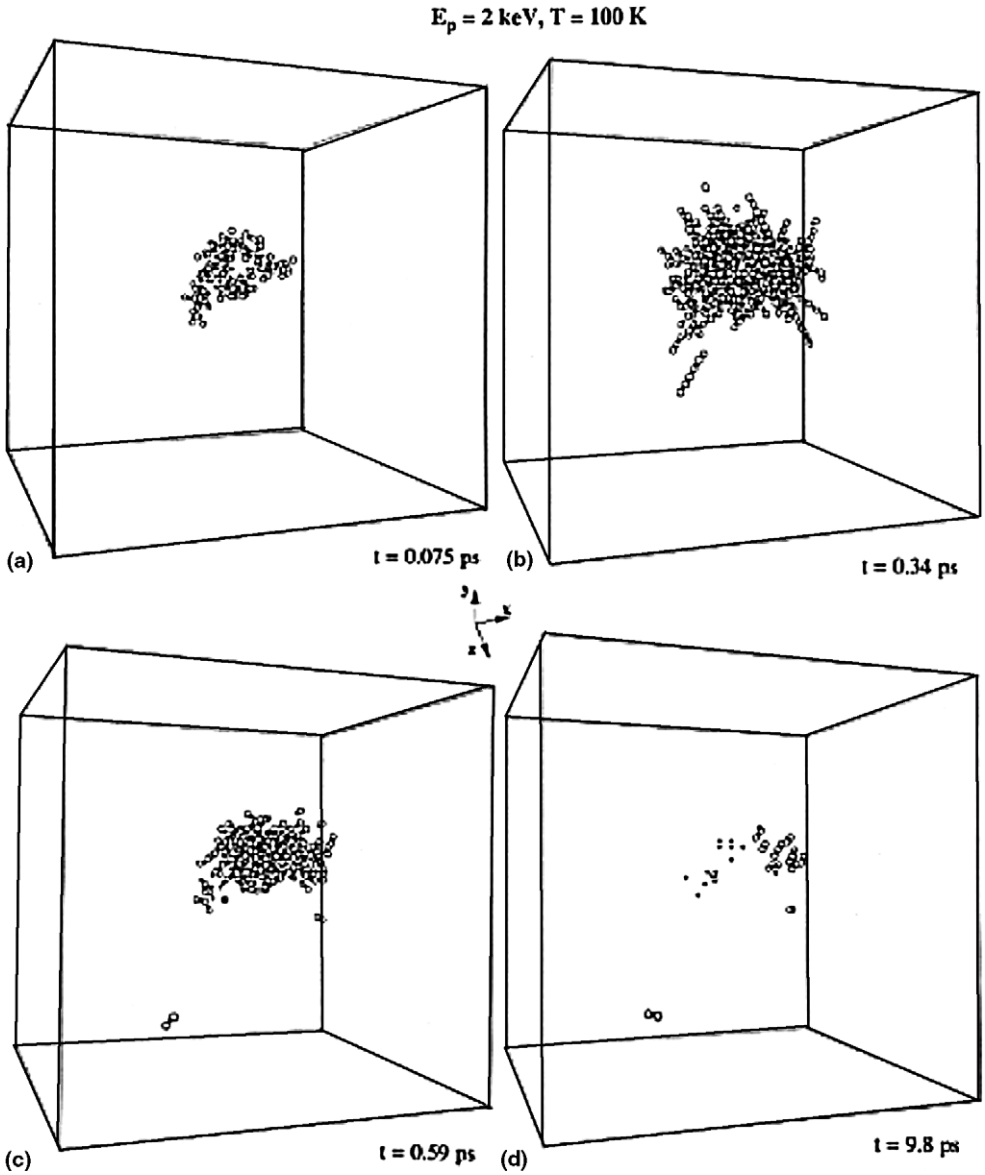

Fig. 2. Molecular dynamics simulation of stages in displacement cascade formation in Fe. Incident particle of energy 2 keV incident at a [1 3 5] direction. Small spheres are vacant sites and large spheres are self-interstitials. The block size is $30a_0 \times 30a_0 \times 30a_0$. $T = 100$ K. (Figure courtesy Drs David Bacon and Andy Calder.)

of interstitials to clusters is so high that the aggregates are stable against thermally activated dissolution. These stable interstitial clusters are defect sinks and would have to be considered in calculation of steady state point defect concentrations. In contrast most of the vacancy clusters decay to feed individual vacancies into the defect sea in the matrix.

The production rate of mobile single vacancies is thus significantly greater than that of mobile interstitials. This asymmetry is known as a 'production bias' [16]. The situation is analogous to that in high-temperature irradiations where thermal and irradiation-induced vacancies are created at similar rates. In both cases a sort of steady state in mobile defect concentrations will be established, wherein the arrival rate vacancies at such neutral sinks as grain boundaries will be significantly greater than that of self-interstitials.

The vacancy aggregates shown in Figs. 1–3 are relatively high energy configurations. Certainly

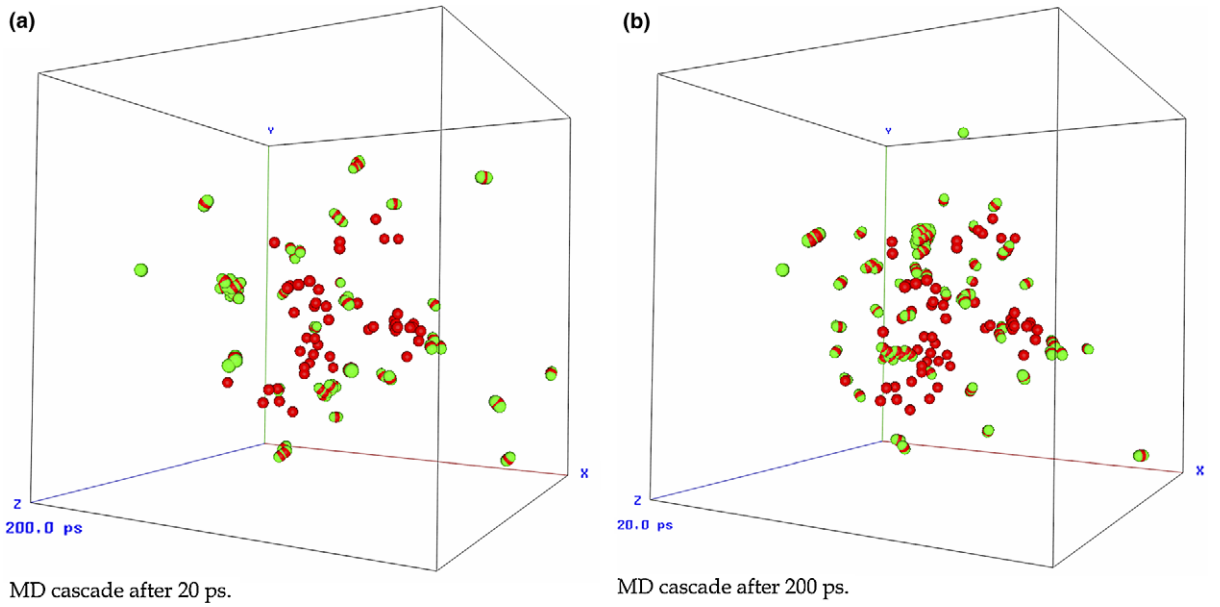

Fig. 3. Views of the 20 keV cascade in Fe at 100 K from MD simulations. Green spheres are displaced atoms, red spheres are unoccupied lattice sites. A self-interstitial atom appears as two displaced atoms (green spheres) sharing one lattice site (red sphere). Vacancies appear as isolated red spheres. Box dimension is ~14 nm on a side. (a) 20 ps, (b) 200 ps. (Figure courtesy Dr R.E. Stoller.)

there will be a tendency for the vacancy strings and diffuse aggregates to agglomerate into compact clusters. Such rearrangement will require times well beyond those available in molecular dynamics.

Wirth [17] used the technique of Monte Carlo simulation to describe the evolution of cascades from the picosecond to millisecond range at a temperature of 563 K. He started with the 20 keV cascade in iron shown in Fig. 3. Interstitials and interstitial clusters were removed before the simulation. Inclusion of these defects would have caused the mono-vacancy concentration to decrease even faster than it did.

Wirth assumed an absorbing boundary condition so that vacancies reaching the boundary were removed from the calculation and not replaced. In this way the system became under saturated with respect to vacancies and all clusters tended to dissolve. Fig. 4(a)–(c) shows the evolution of the vacancies and vacancy clusters.

The peak in number of vacancy clusters occurs at about 70 μs. At this time the cascade has left behind one cluster of a dozen vacancies, and a number of di-, tri-, and tetra-vacancies. Most of the vacancies are in clusters with the remainder left as single vacancies. After this time, the clusters undergo a combination of coarsening and dissolution. The absorbing boundary condition serves to emphasize dissolution. There will in fact be a high supersaturation of vacancies in a real material, which will tend to give cluster growth.

We will take the situation existing at about 70 μs as characteristic of the vacancy clustering occurring in the displacement cascade. Wirth produced a movie of atomic motions during cascade aging that showed large amounts of internal rearrangement within the clusters between vacancy loss events. The vacancy clusters thus may be taken as being internally equilibrated. We will use this information on sizes and numbers of vacancy clusters as input parameters in our calculations.

3. Nucleation theory

Classical nucleation theory [18] is capable of describing void nucleation from a sea of supersaturated vacancies. Katz and Wiedersich [8] and Russell [9] independently extended the theory to include the effects of radiation-induced self-interstitial atoms.

Nucleation is represented [18] by diffusion of a particle along a size coordinate, x, equal to the number of vacancies in the void. Vacancy capture moves the void one step in the positive direction, while vacancy emission or interstitial capture result in movement in the negative direction. The flux of

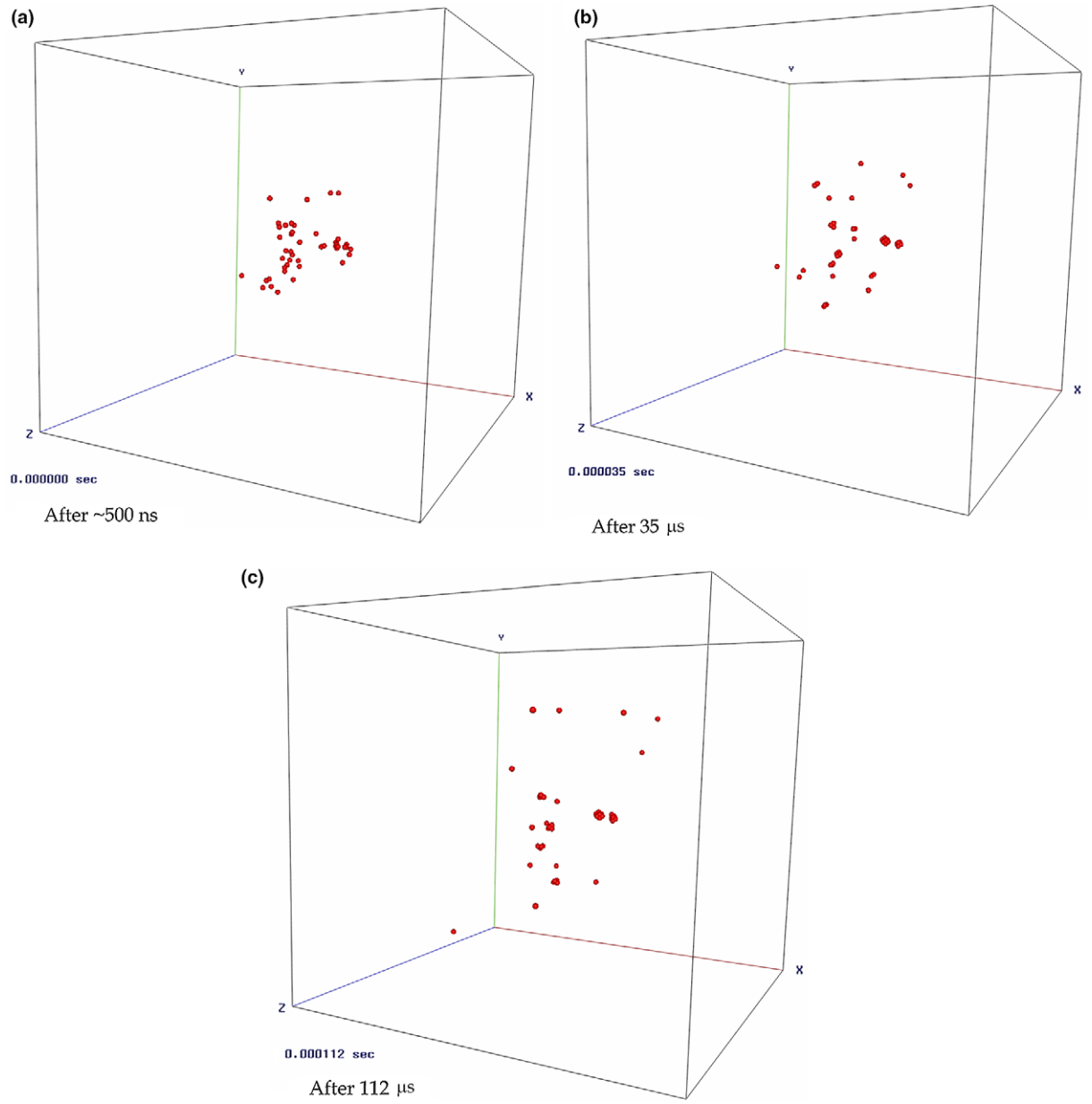

Fig. 4. Results from the kinetic lattice Monte Carlo simulation of the evolution of the cascade in Fig. 3. Self-interstitial atoms and clusters have been removed. Heavy vacancy clustering is seen, followed by coarsening and dissolution. $T = 563$ K. (a) After ~500 ns, (b) after 34 μs, (c) after 112 μs. (Figure courtesy Dr Brian Wirth.)

clusters, J_x, along the size coordinate, x, is given by Russell [9] as

$$J_x = -\beta(x)C^0(x)\left(\frac{\partial C(x,t)/C^0(x)}{\partial x}\right), \quad (1)$$

where

$\beta(x)$	rate of capture of vacancies by x-mer
$C(x,t)$	number of clusters of x vacancies (x-mers) per unit volume
$C^0(x)$	$N\exp(-\Delta G^0(x)/kT)$ equals concentration of x-mer giving $J_x = 0$
kT	Boltzmann factor
N	number of atomic sites per unit volume

$\Delta G^0(x)$ is a kinetically modified potential that plays the same role as the free energy of activation. $\Delta G^e(x)$, in nucleation without self-interstitial involvement.

In the simple capillarity approximation, assuming spherical voids,

$$\Delta G^{e}(x) = xkT\ln S + (4\pi)^{1/3}(3V_a)^{2/3}x^{2/3}\gamma, \tag{2}$$

where V_a = atomic volume, S = ratio of actual and thermal equilibrium vacancy concentrations and γ = interfacial energy between the void and matrix.

The kinetically modified potential is given by

$$\Delta G^{0}(x) = kT\sum_{x=0}^{x-1}\ln[\beta_i/\beta_v + \exp[(\Delta G^{e}(x+1) - \Delta G^{e}(x))/kT]], \tag{3}$$

where β_i/β_v is the ratio of interstitial and vacancy arrival rates at a given size of void. Typically, β_i/β_v is thought to be about 0.9 and must be less than unity for void nucleation or growth to occur at all.

For a spherical void it is easy to calculate the radius of the critical nucleus as

$$r^* = 2\gamma V_a/\{kT\ln[S(1-\beta_i/\beta_v)]\}. \tag{4}$$

The number of vacancies in the critical nucleus is given by

$$x^* = 4\pi(r^*)^3/3V_a. \tag{5}$$

The kinetically modified potential $\Delta G^0(x)$ exhibits a maximum of ΔG^* at the critical nucleus size, x^*: these critical nuclei have an equal probability of growing and decaying and are in a sort of unstable equilibrium with the surrounding matrix. The nucleation process is throttled by the need for clusters to pass over this activation barrier.

Application of the divergence theorem to Eq. (1) gives

$$\frac{\partial C(x,t)}{\partial t} = \frac{\partial}{\partial x}\left\{\beta(x)C^0(x)\left(\frac{\partial(C(x,t)/C^0(x))}{\partial x}\right)\right\}. \tag{6}$$

Eq. (6) may be integrated at steady state, where $\partial C(x,t)/\partial t = 0$, to give the steady state nucleation rate, J^*, in nuclei/m^3/s.

$$J^* = Z\beta^* C^0(x^*), \tag{7}$$

$\beta^*$and $C^0(x^*)$ are $\beta(x)$ and $C^0(x)$ evaluated at critical nucleus size, x^*. In particular

$$C^0(x^*) = N\exp(-\Delta G^*/kT), \tag{8}$$

$$Z = -\left(\frac{\partial(C(x)/C^0(x))}{\partial x}\right)_{x^*}. \tag{9}$$

The Zeldovich factor, Z, accounts for the concentration of critical nuclei being less than the equilibrium, and for the possibility of supercritical nuclei decaying to sub critical size: typically, $Z \approx 0.1$. The frequency factor, β^*, is the impingement frequency of vacancies on the critical nucleus.

The steady state nucleation rate, J^*, is as always [18] dominated by ΔG^*, which in turn is especially sensitive to the value of the interfacial energy, γ. The functional form of $\Delta G^0(x)$ is reflected in the value of Z. The maximum possible value of Z is unity, for a sharply peaked barrier, and the minimum is $\sim 1/2x^*$, for a broad flat barrier. We are dealing with x^* the order of 10, so any uncertainty in Z is of minor consequence compared to those in ΔG^*.

Classical nucleation theory is obtained by taking $\beta_i/\beta_v = 0$. The effect of the self-interstitials is to increase both n^* and ΔG^* and thereby make void nucleation more difficult. Self-interstitial involvement typically reduces the void nucleation rate by several orders of magnitude.

4. Void nucleation with vacancy cluster injection

Wirth and Odette [19] and Caturla et al. [20] report on Monte Carlo simulations of void formation in the presence of displacement cascades. This work is apparently based on the Wirth thesis [17] which used a cell of some one million atoms. In this way they were able to include the details of the cascade structure in the simulation only for a very small volume of material.

Trinkaus and Singh [21] studied void nucleation under cascade damage conditions. Their key quantities were the effective supersaturations for vacancies and gas atoms. They were able to simulate the rapid saturation of void density that is experimentally observed. They did not attempt a full kinetic analysis of void nucleation.

Recently a theory was formulated [22] that considered the effects of embryo injection on the nucleation rate. Clusters are assumed to be injected into the system, at a rate, $Q(x)$, per unit volume, time, and size class.

The continuity equation (3) then becomes

$$\frac{\partial C(x,t)}{\partial t} = \frac{\partial}{\partial x}(\beta(x)C^0(x))\left(\frac{\partial(C(x,t)/C^0(x))}{\partial x}\right) + Q(x). \tag{10}$$

Eq. (10) may be rendered tractable by considering only steady state, taking $y = C(x)/C^0(x)$, and approximating the activation barrier as shown in Fig. 5. We take $G^2 = \frac{\Delta G^*}{kTx^*}$. so that for $x < x^*$, $C^0(x) = N\exp(-G^2x)$, and for $x > x^*$,

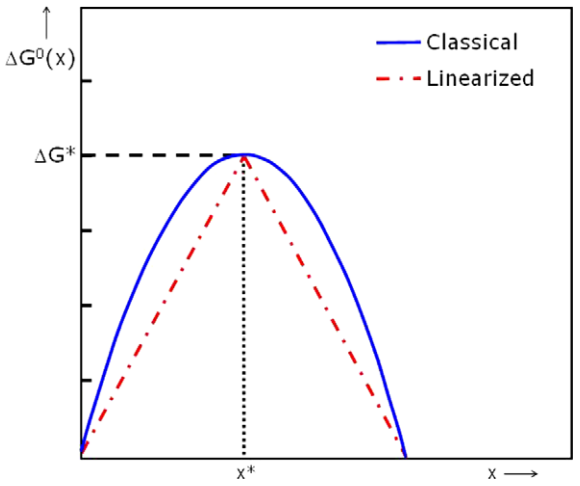

Fig. 5. Comparison of linearized activation barrier to usual curved barrier. The two barriers have the same x^* and ΔG^*.

$$C^0(x) = N\exp[-G^2(2x^* - x)]. \tag{11}$$

As noted earlier the physics of nucleation are such that the most important quantity in J^* is ΔG^*. The precise shape of the barrier is of secondary concern. The linearization preserves ΔG^* (as well as x^*) hence is not expected to introduce serious error in the steady state nucleation rate. We also let $\beta(x)$ take its value at critical nucleus size, β^*, which a minor approximation.

Eq. (10) becomes (for $x < x^*$)

$$\frac{d^2y}{dx^2} - G^2\frac{dy}{dx} + \frac{Q(x)}{\beta^* C^0(x)} = 0. \tag{12}$$

For $x > x^*$ the coefficient of $\frac{dy}{dx}$ changes sign.

The first term in Eq. (12) corresponds to one-dimensional random walk with a unity diffusion coefficient, the second to diffusion in a potential field, and the third to cluster injection. Eq. (12) is a linear, first-degree ODE with constant coefficients, a standard form, and may be solved directly by the method of the Laplace transform or from handbooks [23].

The solution to ODE's such as Eq. (12) is the sum of the complementary function, $F(x)$, and the particular integral $I(x)$ [23]. The complementary function is the solution to the complementary equation of Eq. (12), given by

$$\frac{d^2y}{dx^2} - G^2\frac{dy}{dx} = 0. \tag{13}$$

The solution is

$$F(x) = A_1 e^{r_1 x} - A_2 e^{r_2 x}, \tag{14}$$

where $r_1 = G^2$, $r_2 = 0$. The two arbitrary constants, A_1, A_2, are determined from the boundary conditions. The particular integral contains no arbitrary constants and is given by

$$I(x) = -e^{r_2 x}\int e^{-r_2 x}e^{r_1 x} \times \int e^{-r_1 x}\left(\frac{Q(x)}{\beta^* C^0(x)}\right)(dx)^2. \tag{15}$$

The solution is then

$$y = A_1 e^{G^2 x} - A_2 + \frac{1}{\beta^* N}\int e^{G^2 x}\int Q(x)(dx)^2. \tag{16}$$

The cluster flux through the size classes is determined by the cluster concentrations and the rates of single defect capture or loss. Eq. (7) will thus still give the nucleation rate but with a modified Zeldovich factor obtained from Eq. (16). Knowledge of $\left(\frac{dy}{dx}\right)_{x^*}$ thus allows one to calculate J^*.

Eq. (16) puts limits on the functional form of the source function, $Q(x)$. Not only must $Q(x)$ be integrable, its integral, times $\exp(G^2 x)$ must also be integrable. The choice of source functions is thus sharply limited.

We make the common assumption that the concentrations of the smallest clusters are in equilibrium with the mono-vacancy concentration, such that $y(x)$ approaches unity as x approaches zero. Also, all supercritical clusters are immediately removed from the system so that $y(x^*) = 0$.

A physically plausible source function, $Q(x)$, should have $Q(0) = Q(x^*) = 0$, and show one or more sharp peaks in the interval, $0 > x < x^*$, so that only clusters of a particular sizes are injected. The zero condition at $x = 0$ is to prevent problems in assuming $y(0) = 1$ while requiring $Q(x^*) = 0$ is needed to keep from injecting clusters of critical nucleus size. Such clusters would obviously grow and probably mask the effects of inserted sub critical clusters.

The Dirac delta function and the Gaussian function have the desired properties, but render Eq. (16) non-integrable. The function

$$Q(x) = Q_0\,\mathrm{Sin}^4[(2n+1)\pi x/x^*], \tag{17}$$

where n = integer, peaks fairly sharply, goes to zero rapidly at $x = 0$, x^* and renders Eq. (16) integrable. Taking $n = 0$ corresponds to injection of clusters of sizes about $x^*/2$, and $n > 0$ corresponds to injection of vacancy clusters of several sizes.

Wirth's Monte Carlo simulations showed that there was a range of vacancy cluster sizes in the residue of the displacement cascade. This situation

might be represented in an approximate way by taking $n > 0$ so that there are multiple peaks in $Q(x)$.

There have been too few Monte Carlo simulations of cascade evolution done to give more than a rough idea of the number and sizes of residual vacancy clusters. Our choices of source functions are at best educated guesses.

We were curious as to how much one would have to enhance the flux of clusters into size classes to obtain a significant increase in the nucleation rate. In the absence of cluster injection, $y \approx 1$ for $x < x^*$. We write $J^+(x) =$ flux of clusters from size x to $x+1$. Then approximating $\beta(x) = \beta^*$,

$$J^+ = \beta^* C^0(x) = \beta^* N \exp(-G^2 x). \tag{18}$$

By taking

$$Q(x) = Q_0 N \exp(-G^2 x), \tag{19}$$

it is possible to multiply the non-injection cluster flux at each size class by whatever constant factor we wish. This function for $Q(x)$ also renders Eq. (16) integrable. This calculation is not expected to correspond to any particular physical situation, and is done to give a bit of perspective on what level of cluster injection is needed to give an enhanced nucleation rate.

$Q(x)$ is the number of embryos per unit volume and time per size class. The total number of embryos is Q_T, where

$$Q_T = \int_0^{x^*} Q(x)\mathrm{d}x. \tag{20}$$

It was found that Q_T/Q_0 equaled $1/G^2$ for the exponential injection rate and 30/8 for the trigonometric forms of $Q(x)$.

Our procedure was to evaluate the particular integral for a given $Q(x)$ and then solve for the arbitrary constants, A_1 and A_2. Solving for the arbitrary constants involved some messy equations: those interested may consult the Kim thesis [24].

Table 1 gives the assumed material and irradiation parameters. The irradiation rate is that for fast reactor cladding and the temperature is at the low end of the range of irradiations. Our assumed nucleation parameters were $x^* = 10$, $G^2 = 6$, and $\beta^* = 1/s$. These quantities gave an activation barrier of $\Delta G^*/kT = 60$ and a steady state nucleation rate of $J_0^* = 5 \times 10^3$ voids/m^3/s. The subscript indicates the absence of embryo injection. These parameters were chosen to give a nucleation rate below that actually observed in reactor materials so that the effects of embryo injection would be easily observable, should they occur at all.

Table 1
Material and irradiation parameters

Matrix	FCC metal
Temperature	$T_m/3$
Displacement rate	10^{-6} dpa/s.
ΔG^*	$60kT$
x^*	10 vacancies
J_0^*	5×10^3 voids/m^3/s

5. Results and discussion

The injected embryos had little effect unless $Q_0 > J_0^*$. That is the number of injected embryos had to be greater than the number of embryos achieving critical size in the absence of injection. This is reasonable, as an injection rate of much less than J_0^* would simply be a drop in the bucket compared to the nuclei formed anyway.

We found with the trigonometric source functions that for $Q_0 > J_0^*$, the nucleation rate was about equal to Q_0. which indicates that most of the injected voids were reaching critical nucleus size. We were surprised to find that the nucleation rate was nearly the same for $n = 0$ or $n = 1$. This result is counter to intuition, which says that the closer the injected cluster size is to x^* the larger the effect should be.

The result was different for the exponential form of cluster injection. Values of Q_0 giving huge enhancements of nucleation rate in the other cases had no effect here. This result is somewhat in line with intuition. The exponential decreases so rapidly with x that most of the injected clusters are mono- or di-vacancies, which should not have a profound effect on the nucleation rate.

The results of our calculations should be looked upon as preliminary. We are continuing and rechecking our calculations.

It is illustrative to estimate what neutron irradiation conditions would correspond to a particular embryo injection rate. The molecular dynamic Monte Carlo studies discussed earlier found that a approximately one quarter of the vacancies produced by displacement cascades wind up in clusters in times in the μs–ms range. In one case studied by Wirth [17] clusters as large as a dozen vacancies formed, with larger numbers of smaller clusters. We consider a displacement rate of 10^{-6} displaced atoms/atom/s, or 10^{23} displacements/m^3/s as characteristic of fast reactor cladding materials. Then, Q_T is the order of 10^{21}–10^{22} embryos/m^3/s.

This rate of embryo injection would give a huge increase in the void nucleation rate by any source function except the exponential. The observed void nucleation rates in fast reactor cladding are the order of 10^{15}–10^{20} voids/m^3/s, which is of the same magnitude as the rate due to cluster injection. This very rough correspondence of these numbers lead us to suspect that embryo injection may play an important role in void nucleation during neutron irradiation. This effect would be less significant were nucleation rapid in the absence of embryo injection.

We note that the vacancy clusters formed as residues of cascades must be below the critical nucleus size. Were they larger, they would simply grow in the sea of supersaturated vacancies and nucleation would not be needed to form voids. Many observations of void swelling have shown that nucleation is needed, hence our taking the vacancy clusters as being of sub-critical nucleus size is appropriate. Whether or not a vacancy cluster is of critical nucleus size of course depends crucially on the value of the interfacial energy and on the vacancy concentration in the surrounding matrix.

The preceding calculations must be considered only a first estimate of the effects of cluster injection on the void nucleation rate. Calculations for specific sets of irradiation conditions are needed to be definitive.

6. Conclusions

The effects of embryo injection on the nucleation rate have been analyzed as an extension to existing void nucleation theory.

Sub-critical embryo injection may significantly enhance the rate of void nucleation.

The nucleation rate is surprisingly insensitive to the size of the injected embryos.

Embryo injection may play an important role in void nucleation under cascade-producing irradiation such as occurs in advanced fission and fusion reactors.

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