CASCADE DIFFUSION THEORY OF DISPLACEMENT-INDUCED
POINT DEFECT CONCENTRATION AND FLUX FLUCTUATIONS*L. K. Mansur, A. D. Brailsford¹
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ABSTRACT

A theoretical approach, which has been developed to assess the fluctuations in point defect concentrations and fluxes to sinks that are induced by the production of point defects in spatially and temporally discrete cascades, is summarized. Solutions for cascade dissipation are outlined for the case where cascades occur in a homogeneous lossy medium as well as for the more involved geometries where cascades occur in the presence of a nearby dislocation or cavity. By superposition of solutions representing discrete cascades under conditions of interest, time profiles of point defect concentrations and fluxes are generated. The profiles exhibit extreme fluctuations. Continuum rate theory results arise by applying limits and approximations to cascade diffusion theory. Application of the theory to microstructural processes shows that property changes, especially creep are affected by the fluctuations.

INTRODUCTION

Much of the recent interest in the production of point defects has centered on details of their production in displacement cascades [1]. However, any theoretical analysis of the annealing and ultimate fates of point defects usually is based on continuum rate theory. In this approach point defect production is modeled as uniform in space and continuous in time. In particular, cascade-induced fluctuations in point defect concentrations and fluxes from point to point and time to time are excluded from this analysis by definition. The questions naturally arise as to what physics has been lost in this approximation and what processes might be better understood if a more realistic theory, were available.

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The same type of question arises in the analysis of "pulsed" or discontinuous irradiations. Irradiation pulsing with various periods is thought to be a powerful tool for separating various point-defect-aggregation-driven microstructural development processes according to their relaxation times [2]. There is technological interest in the effects of irradiation pulsing since in fusion reactor engineering some machine designs call for pulsed operations [3]. The first walls of these reactors will be subjected to high flux pulsed neutron irradiation. However, even in a steady irradiation every small local region of material experiences a kind of intrinsic pulsed irradiation from cascades [4]. A new theoretical approach is called for that recognizes intrinsic cascade-induced fluctuations in order to correctly evaluate extrinsic pulsing.

Recently, a cascade diffusion theory has been developed in response to these needs [5,6]. In the present paper we outline the basic formulation, give predicted results for point defect concentrations and fluxes, describe several applications, and show how by suitable approximations and limits the continuum rate theory can be derived from the present more general approach.

CASCADE DIFFUSION THEORY

In the present approach, point defects are introduced in cascades that occur discretely in time and space. The concentrations of point defects at an arbitrary reference point are obtained as the superposition of point defects from cascades that have occurred everywhere in the material at any past time. Point defects are lost both to microstructural features modeled as point sinks distributed uniformly throughout the continuum (lossy medium) and to nearby finite sinks, either cavities or dislocations. Thus, convenient aspects of the conventional rate theory are retained including the continuum diffusion approximation and the construction of a lossy medium to replace the real material containing microstructural sinks. The crucial physical aspect of point defect production in discrete cascades is the new ingredient. Because the presence of nearby finite sinks perturbs the diffusion field of a cascade, the cavity and the dislocation have also been introduced into the lossy medium to give solutions of greater accuracy.

The continuity equation describing point defect diffusion from a unit cascade is taken as [6]

$$\left[\frac{\partial}{\partial t} - D(\nabla^2 - S) \right] g(\underline{r}, \underline{R}; t, t') = \delta(\underline{r} - \underline{R}) \delta(t - t'). \quad (1)$$

Here D denotes the diffusion coefficient and S the microstructural sink strength for the type of point defect in question. The function g is the Green's function describing the concentration³ at a point $\underline{r}$ and time t

³Here and in the remainder of the paper we ignore point defect recombination. Although recombination may be important in many cases, the approximation is adequate to permit comparisons with the conventional rate theory when recombination is also neglected.

resulting from a cascade at point $\underline{R}$ at time t' . The symbol δ denotes the Dirac delta function. The superposition of concentrations corresponding to all cascades that have occurred is given by

$$c(\underline{r}, t) = \int dt' \int d\underline{R} g(\underline{r}, \underline{R}; t, t') G(\underline{R}, t'), \quad (2)$$

where G is the defect production rate.

The boundary conditions for equation 1 determine the form of the cascade kernel g . The point defect concentration is of interest in three distinct cases: 1) far from any sink (or equivalently in an infinite lossy continuum), 2) near a cavity and 3) near a dislocation. The reason for interest in the latter two is that we ultimately wish to evaluate the fluxes to these sinks in order to describe such processes as swelling and creep. Since the presence of the sink perturbs the cascade solution, the fluxes can only be accurately obtained using the solution derived in the presence of the sink.

SOLUTION WITHOUT FINITE SINKS

Here the boundary conditions are $c = 0$ for $t < t'$ and $c \rightarrow 0$ for $r \rightarrow \infty$ for $t > t'$. By solving Equation 1 using Laplace Transform methods it is straightforward to show that the solution is

$$g(\underline{R}; t, t') = \frac{\exp(-R^2/4D\tau) \exp(-DS\tau)}{(4\pi D\tau)^{3/2}}, \quad (3)$$

for $\tau > 0$. Here $\tau = t - t'$ and we have taken our reference point as the origin so that $r = 0$ and $|\underline{r} - \underline{R}| = R$. We may now estimate the current of point defects to a virtual cavity whose center is placed at the origin, while at the same time indicating the need for a finite sink solution. If the cavity of radius r_c is "small", then the difference in concentration from one place to another nearby given by equation 3 should be small. If we were to regard the cavity as in a uniform far field concentration given by $g(R)$ at any instant then the current to it would be (infinitesimal cavity approximation) $I = 4\pi r_c Dg$ [7], giving,

$$I = \frac{r_c}{2(\pi D\tau)^{3/2}} \exp(-R^2/4D\tau) \exp(-DS\tau). \quad (4)$$

However, we need a reference scale in order to say that the cavity satisfies the approximation of being small.

SOLUTION WITH FINITE CAVITY

The solution we seek of equation 1 is that for which the concentration has some fixed value over the cavity surface, say c^{eq} , the thermal equilibrium value. Since we are interested in the irradiation-induced concentration we lose little generality by specifying $c^{eq} = 0$ at $r = r_c$ for all t . This is generally satisfied for the interstitial concentration and it is satisfied

for the vacancies at moderate and low temperatures. Again using Laplace transform methods in a somewhat more involved solution, we find the current to a cavity from a cascade to be

$$I = \frac{r_c(R-r_c)}{2(\pi D\tau)^{1/2}R} \exp\left[-(R-r_c)^2/4D\tau\right] \exp(-DS\tau) \quad (5)$$

for $\tau > 0$. The total number of defects ever collected by a cavity from a single cascade is obtained from equation 5 as

$$n(\infty, t') = (r_c/R) \exp\left[-S^{1/2}(R-r_c)\right] \quad (6)$$

A similar intergration of equation 4 yields a result identical to equation 6 except that R replaces $R-r_c$ in the exponential. Thus the approximation of using equation 4 for the current is valid when $R \gg r_c$ and $S^{-1/2} \gg r_c$. The cavity must be small compared to the distance at which the cascade occurs and small compared to the absorption length of defects in the lossy medium.

SOLUTION WITH FINITE DISLOCATION

Similarly, a solution has been obtained for the cascade in the presence of a nearby dislocation [6]. The solution involves manipulations of double transforms (Fourier, Laplace). One result is that equation 3 can be used to give a good approximation in calculating the current to a dislocation, since $r_\perp$, the dislocation core radius, is generally small with respect to $S^{-1/2}$.

RESULTS

Figure 1 shows characteristic results of the cascade diffusion theory. Shown are calculated time profiles of the vacancy (left) and interstitial concentrations (right) at an arbitrary reference point in a neutron irradiated metal with the relevant parameter values given in Table 1.

Table 1

Parameters used in the calculations (Temperature = 500°C)

Defect Generation Rate (G)	2. x 10 ⁻⁷ defects/atom/s
Sink Strength (S)	1. x 10 ¹⁵ m ⁻²
Vacancy Diffusion Coefficient (D _v)	1. x 10 ¹⁵ m ² /s
Interstitial Diffusion Coefficient (D _i)	8.4 x 10 ⁻⁹ m ² /s

Both profiles show large fluctuations. The interstitial component is more like a series of intense spikes of very short duration separated by relatively long time intervals where no interstitial concentration exists, while the vacancy profile consists of a relatively steady background punctuated by large spikes. The differences between the vacancy and interstitial profiles are due entirely to the difference in diffusion speed; the interstitial is so fast that the concentration at a point is virtually never contributed by more than one cascade, while the diffusion speed of the vacancy is such that approximately 10^3 cascades contribute defects at the reference point in the steady state.

In the usual rate theory picture the concentration is given in the steady state by $C = \frac{G}{DS}$, yielding a horizontal line on Figure 1 at $2 \times 10^{22} \text{ m}^{-3}$ for the vacancy and about 2.4×10^{15} for the interstitial. Figure 1 is generated with the solution given by equation 3. This is also an approximation to the current profile to a cavity when multiplied by $4\pi r_c D$, equation 4. Figure 2 shows a comparison of results generated using this approximation with results obtained from the exact solution, equation 5, for a cavity of 20 nm radius. There are large differences in the instantaneous current as well as the total current.

This analysis also enables us to evaluate the importance of each increment of material volume in contributing point defects to the concentration at the reference point. It is easy to show from equations 2 and 3 that the average concentration contributed by cascades in the material between reduced radii X_1 and X_2 , where $X = R/S^{-1/2}$, is

$$C_{\Delta} = \frac{G}{DS} \{ [\exp(-X_1) - \exp(-X_2)] + [X_1 \exp(-X_1) - X_2 \exp(-X_2)] \}. \quad (7)$$

A similar result is also available for the incremental contributions of each region to the current to the current to a cavity. From equation 7, spatial importance functions may be obtained.

For example, if we set $X_1 = 0$ and $X_2 = X$ we may evaluate both the contribution from all of the material within X [equation 7] and the contribution of each infinitesimal shell of material at distance X [derivative of equation 7]. These results are shown in figure 3. We note that the most important material contributing defects lies at the radial distance $X = 1$ ($R = S^{-1/2}$), and that about 80% of the defects comprising the concentration at an arbitrary reference point come from material within three absorption lengths ($3S^{-1/2}$).

Although cascades throughout the material contribute to the local concentration in principle, virtually no defects come from distances greater than $7S^{-1/2}$.

Figure 1. Calculated time profile for the vacancy (left) and interstitial (right) concentrations using the parameters shown in Table 1.

Figure 2. Comparison of the current profiles to a cavity of size 20 nm resulting from steady irradiation using the exact solution (left) compared with the results of the infinitesimal cavity approximation (middle) and the difference between the two (right).

Figure 3. Spatial importance functions for the average defect current to a cavity. Upper plot shows the differential function, and lower plot shows the integral function.

APPLICATIONS

The present theoretical approach is applicable to a wide variety of investigations. In some cases it is intuitively obvious that the present approach and the continuum rate theory approach should give the same results. For example, consider the growth of large cavities. Whether vacancies and interstitials are accumulated in widely fluctuating fluxes or are accumulated at the corresponding time-averaged rate, it is the net cumulative difference of vacancy over interstitial absorption that is important in the growth of large cavities. Thus the cascade diffusion theory and the continuum rate theory give the same results in this case, except for small non-linear effects [5]. These non-linearities arise from the fact that shrinkage induced by a large fluctuation in interstitial absorption requires the cavity to emit thermal vacancies more rapidly at its smaller size -- the emission rate is exponential in the cavity radius. When next the cavity absorbs the excess vacancies that it must receive to keep the overall point defect fluxes at their average values, the cavity now can grow only to a smaller size than it would have if interstitials and vacancies had been absorbed continuously at their average rates.

In other cases, however, the presence of fluctuations may be crucial and more detailed investigation is warranted. Two cases that we have examined are 1) the possible enhanced transition of small cavities from subcritical gas-stabilized bubbles to cavities that grow inexorably by excess vacancy absorption, and 2) the existence of a new mechanism of irradiation creep that is caused by cascade-induced point defect fluctuations.

CASCADE-INDUCED CAVITY TRANSITIONS

The existence of a critical radius and corresponding critical number of gas atoms as a requirement for cavity growth by excess vacancy absorption is now well established [8]. If the cavity contains a number of gas atoms n_g that is less than a critical number of gas atoms, n_g^* , then the critical radius for bias driven cavity growth r_C^C is larger than the minimum critical radius r_C^* . (If the cavity contains more than n_g^* gas atoms then there is no critical radius and the cavity will always grow by bias-driven growth.)

When $n_g < n_g^*$ the cavity resides at a stable radius $r_C^S < r_C^C$. It can only achieve bias-driven growth if by fluctuations it makes an excursion from radius r_C^S to a radius as large as or larger than r_C^C . If there are no fluctuations then the cavity must wait until it absorbs n_g^* gas atoms before it begins bias-driven growth. Our inquiry here centers on whether cascade-induced fluctuations can be significant enough to cause an early transition from r_C^S to r_C^C , without the necessity of waiting to accumulate n_g^* gas atoms. The cavity is only likely to be converted when the magnitude of cascade

induced fluctuations is comparable to the number of vacancies required to produce a cavity of radius r_C^C . Our detailed analysis shows that there is some effect on producing early transitions but that this is probably not important in the context of normal experiments on radiation-induced swelling [9]. Figure 4 shows the calculated results. The parameter values are given in reference 9. While the temperature regime where this behavior occurs depends on the parameter values, the general features do not. At low temperatures, where r_C^S and r_C^C are both small, the effect of fluctuations on causing early transition is relatively large. However, at low temperatures the incubation dose is very short anyway because little gas is required to be accumulated for such small critical radii. In most cases of interest, therefore, a reduction in dose from the order of one dpa to a fraction of a dpa would probably not be important. At high temperatures the sizes of r_C^S and r_C^C are so large that cascade-induced fluctuations only make a negligible difference in the transition dose.

Figure 4. Leftmost curve shows fractional change in the onset of swelling due to cascade-induced fluctuations. Upper curve is incubation dose obtained neglecting fluctuations while lower curve includes fluctuations.

CASCADE-INDUCED CREEP

A dislocation segment pinned on an obstacle can undergo glide and contribute to creep deformation if it can be unpinned. Climb can unpin it and there are several theoretical models of creep by climb-enabled glide. In these, climb is gradual and cumulative -- a given dislocation climbs only in one direction driven by excess absorption of point defects of one type. In swelling-driven creep [10] each dislocation absorbs the excess interstitials corresponding to the vacancies in cavities. In preferred absorption glide creep [11] a dislocation receives either excess vacancies or interstitials depending on the relative orientation of its Burgers vector to the stress axis.

With reference to figure 1 a new mechanism of irradiation creep can be visualized [12]. In response to the cascade-induced fluctuations in point defect fluxes, a dislocation segment will make climb excursions in both directions. During an excursion there is a probability that the segment will reach an unpinning position on the obstacle and be freed to glide. The calculation of this unpinning frequency and subsequent creep deformation is possible in the present theory.

It has been found that the unpinning frequency is substantial when compared to that caused by swelling and by stress induced preferred absorption. Table 2 shows the calculated unpinning frequencies for a dislocation segment calculated for cascade-induced creep, swelling-driven creep, and stress-induced preferred absorption glide processes. The results are shown as unpinning frequencies rather than creep rates because the creep rates would be in the same ratio regardless of the model used to translate the unpinning and glide into creep. For obstacles of height $40b$ it is found that cascade-induced creep gives the largest contribution for the parameter values of reference 12. However, for increasing obstacle size the relative contribution of cascade induced creep diminishes, and for obstacles twice this height, swelling driven creep becomes the dominant mechanism.

Table 2

Release frequencies of a dislocation segment
from obstacles of height $40b$

<u>Creep Mechanism</u>	<u>Release Frequency (s^{-1})</u>
Cascade-Induced	4×10^{-5}
Swelling-Driven	4×10^{-6}
Preferred-Absorption	1×10^{-7}

DISCUSSION AND CONCLUSIONS

Under appropriate limits and approximations the conventional rate theory can be obtained from the cascade diffusion theory. For example, if we derive the mean value of point defect concentration from the present theory this should correspond to the point defect concentration of the conventional rate theory. From equation 3 we find that the mean concentration $C(t)$ far from sinks is

$$C(t) = 4\pi G \int_0^t dt \int_0^\infty \frac{\rho^2 \exp[-\rho^2/4D\tau] \exp(-DS\tau)}{(4\pi D\tau)^{3/2}} d\rho \quad (8)$$

giving

$$C(t) = (G/DS)[1 - \exp(-DS t)]. \quad (9)$$

This expression, equation 9, is a standard result of rate theory [13]. At large times the result is $C(\infty) = G/DS$, a result often used to calculate the concentration when recombination is neglected. In a similar way we derive the mean value of the current to a cavity from all prior cascades in all space from equation 4 as

$$I(\infty) = (4\pi r_c G/S)(1 + S^{1/2} r_c). \quad (10)$$

Again this is a standard result of continuum rate theory. We note that the effect of using the Green's function that recognizes the cavity's presence, equation 4, is simply to introduce the multiple sink interaction factor $S^{1/2}r_c$ [7] into equation 10. The corresponding result for the dislocation is

$$I(\infty) = \frac{2\pi S^{1/2} r_{\perp} K_1(S^{1/2} r_{\perp}) G}{SK_0(S^{1/2} r_{\perp})} \quad (11)$$

Here the point defect current is per unit line length of dislocation, the symbol K_j denotes the Modified Bessel Function of the second kind of order j , and the symbol $r_{\perp}$ denotes the dislocation capture radius.

To summarize, cascade diffusion theory provides a more faithful representation of the point defect concentrations and fluxes in an irradiated material than does continuum rate theory. In particular the details of the extreme cascade-induced fluctuations can be described quantitatively. In turn the analysis of processes that may depend on fluctuations rather than on averages or on long term cumulative fluxes is made possible. Two such processes are cascade-induced creep and cascade-induced transitions of cavities from the stable to the bias-driven state. On the other hand, for processes that only depend on average quantities, the conventional rate theory is adequate. It is shown that in the appropriate limits the cascade diffusion theory reduces to the conventional rate theory. These results also provide a more rigorous origin for continuum rate theory. Possible further areas to be explored are the nucleation of interstitial clusters and the initiation of phase changes during point defect concentration fluctuations.

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