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EFFECTS OF COLLISION CASCADES ON RECOMBINATION AND CLUSTERING OF POINT DEFECTS

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Abstract

In order to understand the radiation effects under cascade damage conditions where vacancies and interstitials are created in a localized and segregated fashion, a programme has been initiated to model the evolution of point defect microstructure between cascades and among subcascades. The model and some preliminary results are presented of analytical and numerical calculations of the space and time dependent diffusion and clustering of self-interstitial atoms. The calculation includes the effect of back diffusion of self-interstitial atoms to the vacancy-rich cascade core. It is concluded that a diffusion-rate theory model of the point defect evolution in cascade zones appears to be feasible.

1. Introduction

The effects of damage rate and recoil energy spectrum on the microstructural evolution and macroscopic property changes under cascade damage conditions cannot, in our view, be estimated without considering the details of interstitial clustering in cascade zones. It is well known that the distributions of vacancies and interstitial atoms in a nascent cascade* are very localized and segregated [1-3]. Generally, interstitials are found in higher concentrations at the periphery of the cascade zone, while vacancies are concentrated towards the centre of the cascade zone. At temperatures where the interstitials are mobile, the high concentration would unavoidably lead to (a) back diffusion of interstitials into the vacancy-rich cascade zone, (b) formation of interstitial clusters of different sizes and (c) escape of interstitials from the cascade zone. The interstitials escaping the cascade zone would interact with interstitials escaping from other cascades, and interstitial clusters of different sizes would be formed between cascade zones via homogeneous nucleation mechanisms. The growth of these clusters and of those in the cascade zone could lead to the formation of a dislocation network. Additionally, it has been shown recently that the clustering of interstitials in the cascade region causes a "production-bias" [4]. This may provide a major driving force for swelling under cascade damage conditions at the peak swelling temperature.

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* i.e., after the energy spike-induced athermal rearrangement of atoms (the "cooling phase"); approximately 10^{-11} sec

Thus, it is clear that the global microstructural evolution cannot be understood properly without considering the details of interstitial clustering in the cascade zone, as well as between the cascades and among subcascades. However, until now both experimental and theoretical investigations of cascade characteristics have concentrated mostly on studying the behaviour of only the vacancy-rich region of the cascade zone (see e.g. [5-6]).

In the following, we report some preliminary results of an attempt to develop a diffusion-based methodology to study the space- and time-dependent recombination and clustering of point defects generated during the cascade process. The reason for adopting this diffusion-based continuum approach is that global microstructural evolution is a problem too large to be treated in terms of interactions of individual atoms. The discrete treatment of single cascades and subcascades cannot be extended to the global problem without taking into account their interactions. The results obtained using our continuum model should then provide a useful guideline for utilizing the rate theory approach for studying the long term radiation effects under cascade damage conditions. The first step in developing the model is to consider a single cascade.

2. Model

For a single cascade we assume that the vacancies are concentrated at the cascade centre. In the present calculations this vacancy-rich region is simulated by a spherical hole of radius r_c (fig. 1), which is assumed to remain constant. The initial interstitial distribution is assumed to consist of mono-interstitials and interstitial clusters. The

mono-interstitials are concentrated in a spherical shell of radius r_o (fig. 1) surrounding the vacancies with a normal-like distribution defined by a specified standard deviation. The interstitial clusters are treated as dislocation loops and have a specified radius and are concentrated in the same shell with the same distribution as the mono-interstitials. These interstitial loops are assumed to have formed during the cooling phase of the cascade. For the time scale and temperature involved, the mobilities of the vacancies and the tri-interstitials are neglected.

We denote the diffusivities of the mono- and di-interstitials by D_1 and D_2, respectively. Furthermore, the tri-interstitials are considered to be immobile and are treated as interstitial loop nuclei (i.e., initially all loops are tri-interstitials). The disposition of the initial distribution of point-defects in a cascade is depicted in fig. 1.

We consider the following point defect reactions

$$i_1 + i_1 \xrightarrow[\alpha_{11}]{} i_2 \text{ (formation of di-interstitials)} \tag{1}$$

$$i_1 + i_2 \xrightarrow[\alpha_{12}]{} i_3 \text{ (formation of tri-interstitials)} \tag{2}$$

$$i_1 + L \xrightarrow[k_L^2 D_1]{} \text{loop growth} \tag{3}$$

$$i_2 + L \xrightarrow[k_L^2 D_2]{} \text{loop growth} \tag{4}$$

where L represents all the loops, preexisting plus newly formed ones, α_{11}, α_{12}, $k_L^2 D_1$ and $k_L^2 D_2$ are the respective reaction constants. Here k_L^2 is the dislocation loop sink strength.

For this paper, we have neglected the reaction between the di-interstitials. The reaction constants we used are the ones commonly used in the diffusion-limited reaction rate theory [7] assuming that the effects of the inhomogeneity of the point-defect concentrations can be neglected.

The diffusion rate equations for the mono-, di- and tri-interstitial concentrations C_1, C_2 C_3, as well as the dislocation-loop radius R_2 can be written as follows:

$$\frac{dC_1}{dt} = D_1\nabla^2C_1^2 - 2\alpha_{11}C_1^2 - \alpha_{12}C_1C_2 - k_1^2D_1C_1 \tag{5}$$

$$\frac{dC_2}{dt} = D_2\nabla^2C_2^2 + 2\alpha_{11}C_1^2 - \alpha_{12}C_1C_2 - k_2^2D_2C_2 \tag{6}$$

$$\frac{dC_3}{dt} = \alpha_{12}C_1C_2 \tag{7}$$

$$\frac{dR_L}{dt} = Z_1D_1C_1/b + 2Z_2D_2C_2/b \tag{8}$$

subject to the following initial and boundary conditions:

$$C_1(r,o) = C_{10}(r) \tag{9}$$

$$C_2(r,o) = 0 \tag{10}$$

$$C_3(r,o) = C_{30}(r) \tag{11}$$

$$C_1(r,t) = C_2(r,t) = C_3(r,t) = 0 \text{ at } r=r_c \text{ and } r=\infty \tag{12}$$

In equation (8) Z_1 and Z_2 are the dislocation bias factors for mono- and di-interstitials, respectively. Note here that k_1^2 and k_2^2 are both

functions of C_3. Also, the variables C_1, C_2, C_3 and R_L are all functions of both r and t. By discretising in space and using the Green's function method, this initial-boundary-value problem can be converted into a system of 4N first order differential equations that can be integrated numerically. Here N is the number of discrete spatial grid points.

3. Results and Discussion

When the interactions among the interstitials are neglected, the diffusion equation for the mono-interstitials can be solved analytically, a case we treat first. The concentration profiles for various times are calculated and the results are shown in Figure 2 for an initial distribution of 500 interstitials with $r_c = 3b$ and $r_o = 30b$ (b = mono-interstitial jump distance). It can be seen how interstitials are lost via back diffusion to the central hole where they are annihilated. The escape of interstitials away from the cascade region can also be clearly seen in Figure 2.

By allowing the point-defect reactions in equations (1) to (4) to occur, the same initial conditions lead to the results shown in Figure 3. Here, in the region where the interstitial concentration is the highest, clustering of interstitials into immobile tri-interstitials occurs readily. Annihilation through back diffusion and escape from the cascade region also occurs. In the particular situation under consideration, after 1000 jumps, about 50% of the interstitials recombine with the vacancies, 20% cluster, and 30% escape. These fractions are sensitive, as expected, to the initial vacancy-interstitial separation r_o the standard deviation of the distribution and the initial number of interstitials. This reflects the

dependence of recombination and clustering on the details of the cascade morphology.

4. Conclusions

Preliminary results show that a diffusion-rate theory model of the point defect evolution in cascade zones appears to be feasible. Further development of the present model and comparison with results from other approaches, such as molecular dynamics, are necessary to confirm the usefulness of this scheme.

In the present approach, statistics are inherently included and a larger space-time regime can be treated within reasonable computer time.

The validity of the law of mass action in representing the reaction rates in the present calculation must be addressed.

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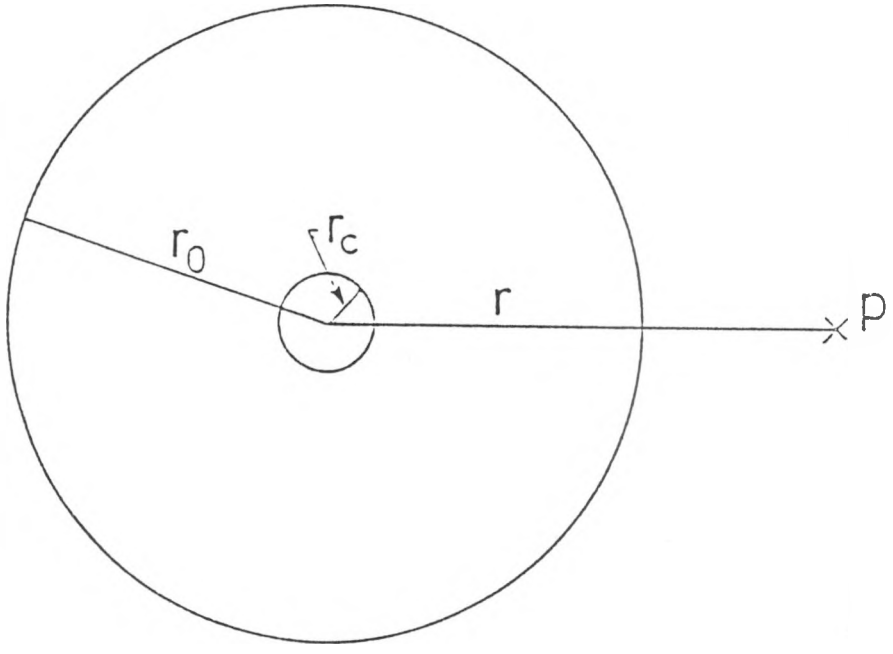
Figure 1: Disposition of the initial configuration of the cascade, consisting of a central vacancy-rich region simulated by a hole and an interstitial-rich region simulated by a shell of an average radius r_o encapsulating the central hole of radius r_c. r defines the point at which the concentration is to be calculated as a function of time.

Figure 2: Evolution of point-defect distributions in a cascade in which mono-interstitial clustering into di-interstitials is neglected. N is the number of single interstitials contained per unit thickness in a spherical shell of radius r which measures the distance (in units of the mono-interstitial jump distance, b) from the cascade centre. The time is measured in terms of the number of mono-interstitial jump.

Figure 3: Evaluation of point-defect distributions in a cascade. (b) and (c) refer to the total number of interstitials remained in the form of di- and tri-interstitials, respectively. Notations are the same as in Figure 2.


r_0
r_c
r
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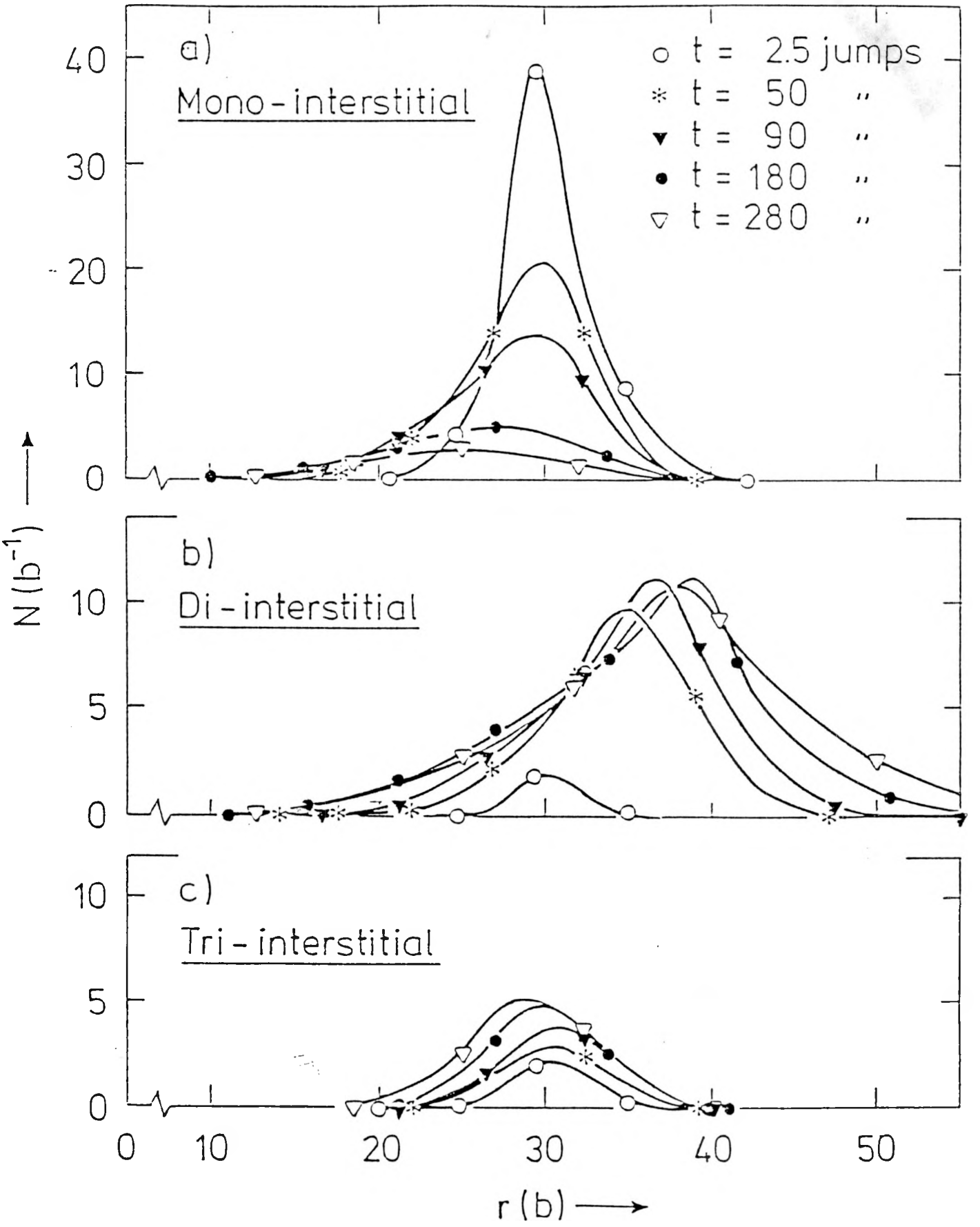
a)
Mono-interstitial
t = 2.5 jumps
t = 50 "
t = 90 "
t = 180 "
t = 280 "
b)
Di-interstitial
c)
Tri-interstitial
N (b⁻¹)
r (b)
0
10
20
30
40
50
5