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A RATE THEORY APPROACH TO TIME DEPENDENT MICROSTRUCTURAL DEVELOPMENT DURING IRRADIATION

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A fully dynamic rate theory for modeling the behavior of point defects in metals during time dependent irradiations has been formulated. The state variable approach has been used to determine the microstructure and defect concentrations as a function of time. The resulting system of stiff non-linear first order ordinary differential equations is solved by the GEAR Program and incorporated into a FORTRAN Computer Code, TRANSWELL. This code has been developed to solve the previous system of equations under a variety of irradiation conditions. It is shown that the point defect and microstructural behavior during the transient phase of an irradiation is quite different than that found after steady state operation.

1 INTRODUCTION

Radiation damage in metallic structures has been studied since the 1940's, and more recently (1966) the phenomenon of void swelling has been added to the long list of mechanisms which may reduce the useful lifetime of fast reactor cladding and fusion reactor blanket structures. Many experimental and theoretical studies have been conducted in the past 10 years on the growth of such voids, and a general understanding of the physics of this phenomenon has been developed for steady-state irradiation by Bullough, Brailsford, and co-workers[1–3] through the rate theory approach. One main feature of the previous work is that the vacancy and interstitial defect concentrations (C_v and C_i, respectively) were assumed to be constant during the irradiation. However, we know that there is a very complex relationship between the development of a dislocation—void microstructure and the defect concentration. The assumption of constant C_v and C_i values is not adequate during the beginning of irradiation when the dislocation microstructure is developing rapidly or during a time dependent irradiation such as that which might occur in pulsed laser fusion reactors.[4] Supporting these observations is the experimental evidence that time dependent heavy ion irradiations can lead to microstructure quite different than those produced under steady-state conditions.[5] Preliminary investigations by Schiffgens and Doran[6] have also shown that there can be a considerable difference between pulsed and steady-state developed microstructure in metals.

The object of this paper is to present a theory and a method of solution that will allow a calculation to be made of the time dependent defect concentrations and microstructural development in metals. This Fully Dynamic Rate Theory (FDRT) relies heavily on the previous work of Bullough[1,3] and extends into a regime which will be of interest to scientists interested in non-steady-state irradiations. For completeness here we will also include the effect of superimposed stress on the behavior of the dislocation components. The FDRT can also be used to predict the microstructure and defect concentrations in a wide variety of metals over a wide range of steady-state irradiations as well.

After a brief description of the general approach to be taken here, we define the parameters used as input to the FDRT. Once the complete set of

coupled equations is developed we describe the method of solution and apply the results to the early stages of a steady-state irradiation and also those conditions which might prevail in during the neutron irradiation of a laser fusion first wall.

2 GENERAL APPROACH

It has been found that one can greatly simplify the mathematical complexities of a dynamic system by describing the kinetic behavior of that system with vector notation. This leads to a state variable approach which is briefly described below. In general, the state of a metal during irradiation can be described by a vector **Y** whose components are defined as follows:

$Y(1)$ = average void radius in cm, r_c
$Y(2)$ = average nonaligned interstitial loop radius in cm, r_{il}^n
$Y(3)$ = concentration of nonaligned vacancy loops per cm^3, N_{vl}^n
$Y(4)$ = concentration of single vacancies tied up in nonaligned vacancy loops in at./at., $q_v{}^n$
$Y(5)$ = total single vacancy concentration in at./at., C_v
$Y(6)$ = total single interstitial concentration in at./at., C_i
$Y(7)$ = average aligned interstitial loop radius in cm, r_{il}^a
$Y(8)$ = concentration of aligned vacancy loops per cm^3, N_{vl}^a
$Y(9)$ = concentration of single vacancies tied up in aligned vacancy loops in at./at., $q_v{}^a$
$Y(10)$ = network creep strain in cm/cm, e

A loop is defined to be aligned if the stress is perpendicular to the plane of the loop.

3 ANALYSIS OF THE STATE VARIABLES

Following the notations of previous work by Brailsford and Bullough[1] and Ghoniem and Kulcinski[7] more detailed expressions for the components of the Y vectors are now derived. First the dislocation densities must be determined and then the vacancy emission rates, defect time constants, defect removal rates, vacancy loop behavior, and finally swelling can be calculated. In order to obtain a more physical relationship with the events taking place in the lattice we will replace the vector notation with symbols more familiar to the reader and then return to the vector notation at the point where a mathematical solution is effected.

3.1 Dislocation Densities

Dislocations in the bulk of a metal can be produced by at least three different mechanisms:

1) Cold work of deformation of the metal which results in initial dislocation network, ρ_d^s.

2) Interstitial loops nucleated during the early stages of irradiation, a fraction of which are preferentially aligned with the applied stress. Their subsequent growth and coalescence form a dislocation network.

3) Vacancy loops are assumed to form athermally in the vacancy rich region of the collision cascade[3] with the initial radius, $r_{vl}(0)$. Their existence, which is also affected by applied stress, is transient in nature because they have a bias to attract interstitials and they thermally emit vacancies at the same time.

If P is the Frenkel pair production rate (dpa/sec), then εP is the fractional rate at which vacancies are removed from solution to form vacancy loops. From conservation of vacancy volume within the loops, the fraction of vacancies tied up in vacancy loops is equal to the volume of a single vacancy loop multiplied by their number density, thus:

$$q_v{}^n = \pi b (r_{vl}^n)^2 N_{vl}^n \tag{1}$$

for nonaligned vacancy loops, and

$$q_v{}^a = \pi b (r_{vl}^a)^2 N_{vl}^a \tag{2}$$

for aligned vacancy loops where b is the Burgers vectors and r_{vl}^n and r_{vl}^a refer to aligned and nonaligned vacancy loop radii, respectively.

For interstitial loops, the dislocation densities are expressed as:

$$\rho_d^{iln} = 2\pi r_{il}^n N_{iln} \tag{3}$$

$$\rho_d^{ila} = 2\pi r_{il}^a N_{ila} \tag{4}$$

while for vacancy loops:

$$\rho_d^{vln} = 2\pi r_{vl}^n N_{vl}^n \tag{5}$$

$$\rho^{vla} = 2\pi r_{vl}^a N_{vl}^a \tag{6}$$

where the following is defined:

ρ_d^{iln} is the nonaligned interstitial dislocation loop line density, cm^{-2}.

N_{iln} is the nonaligned interstitial dislocation loop concentration, cm^{-3}.
ρ_d^{ila} is the aligned interstitial dislocation loop line density, cm^{-2}.
N_{ila} is the aligned interstitial dislocation loop concentration, cm^{-3}.
ρ_d^{vln} is the nonaligned vacancy loop line density, cm^{-2}.
ρ_d^{vla} is the aligned vacancy loop line density, cm^{-2}.
ρ_d^{s} is the deformation produced straight dislocation line density, cm^{-2}.
ρ_d is the total dislocation line density produced by both deformation and radiation, cm^{-2}.

From Eqs. (1) and (2), Eqs. (5) and (6) can be expressed as:

$$\rho_d^{vln} = 2\sqrt{\pi N_{vl}^{n} q_v^{n}/b} \tag{7}$$

$$\rho_d^{vla} = 2\sqrt{\pi N_{vl}^{a} q_v^{a}/b} \tag{8}$$

The total dislocation density in the metal is the sum of all previous components;

$$\rho_d = \rho_d^{s} + \rho_d^{iln} + \rho_d^{ila} + \rho_d^{vln} + \rho_d^{vla} \tag{9}$$

3.2 Vacancy Emission From Microstructural Components

3.2.1 Vacancy emission from voids At high temperatures, the probability of emitting a vacancy from a void becomes appreciable. A larger void surface tension enhances vacancy emission while the presence of gas atoms inside reduces the probability for vacancies to "boil off" the surface of voids. A mathematical description of this process is given as:

$$P_C^{e} = 4\pi r_c N_C D_v C_v^{e} \exp\left\{\left(\frac{2\gamma}{r_c} - P_g(r_c, N_g)\right) b^3/kT\right\} \tag{10}$$

where

P_C^{e} is the fractional vacancy emission rate from the surface of voids, s^{-1}.
N_C is the temperature dependent void concentration, cm^{-3}.
D_v is the temperature dependent vacancy diffusion coefficient, $cm^2\ s^{-1}$.
C_v^{e} is the fractional equilibrium vacancy concentration, at./at.
γ is the surface energy of the void surface, eV/cm^2.
P_g is the gas pressure inside the void, eV/cm^3.
N_g is the number of gas atoms in a void.

Equation (10) is easily derived by considering the vacancy concentration at the voids, C_v, as given by;

$$C_v/C_v^{e} \approx \exp\left\{\left(\frac{\partial F}{\partial n}\right)/kT\right\} \tag{11}$$

where $\partial F/\partial n$ is the change in the configuration energy per vacancy emitted, and C_v^{e} the equilibrium concentration of vacancies. For a spherical hole in an infinite isotropic solid, $(\partial F/\partial n)$ involves the surface energy of the void, the elastic strain energy in the surrounding metal, the applied hydrostatic stress, and the pressure caused by trapped gas atoms inside.

Generalizing the analyses given by Volin and Balluffi[8] and Westmacott, *et al.*,[9] one can write the following expressions.

$$\frac{\partial F}{\partial n} = F_m \Omega \tag{12}$$

where Ω is the atomic volume. Here F_m is the mechanical force per unit surface area acting on a vacancy at the void surface.

$$F_m = P + \frac{2\gamma}{r_C} + \frac{\gamma^2}{2\mu r_C^{2}} - P_g \tag{13}$$

Here P is the hydrostatic pressure, γ the surface energy, r_C the void radius, μ the shear modulus, and P_g the gas pressure.

The gas pressure is always expressed in terms of the number of gas atoms and void radius. If the perfect gas law is used, one gets:

$$P_g = \frac{3NkT}{4\pi r_C^{3}} \tag{14}$$

While if Van der Waals law is assumed to hold, one gets:

$$P_g = \frac{NkT}{(4/3\,\pi r_C^{3} - aN)} - \frac{N^2 d}{16/9\,\pi^2 r_C^{6}} \tag{15}$$

where a and d are constants, and N is the number of gas atoms.

Normally, the elastic energy is negligibly small[8] so in the general case we have:

$$F_m = P + \frac{2\gamma}{r_C} - P_g \tag{16}$$

This formulation is useful in studying the general situation where gas atoms are trapped in voids and where stress waves accompany the damage production.

Although the principal concern in modeling void behavior has been void growth during irradiation, some consideration was given to the response of voids during high temperature annealing.[10] In the absence of irradiation, annealing is of interest for the practical reason that CTR first-wall materials of inertial confinement fusion reactors will be subject to short periods of intense irradiation followed by relatively long periods at high temperature but under no irradiation.

3.2.2 Vacancy emission from deformation produced dislocations For simplicity of calculation, all deformation produced dislocations are assumed to be of the edge type. Edge dislocations that are aligned with the stress have enhanced vacancy emission rates as described below:

$$P_d^{ea} = Z_v^a D_v C_v^e \exp(\sigma\Omega/kT)\rho_d^a \tag{17}$$

$$P_d^{en} = Z_v^n D_v C_v^e \rho_d^n \tag{18}$$

$$P_d^e = P_d^{ea} + P_d^{en} \tag{19}$$

where

P_d^{ea} is the fractional rate of vacancy emission from aligned straight dislocations, s^{-1}.
P_d^{en} is the fractional rate of vacancy emission from nonaligned straight dislocations, s^{-1}.
P_d^e is the total fractional rate of vacancy emission from straight dislocations, s^{-1}.
σ is the uniaxial externally applied stress, eV/cm^3.
ρ_d^{ea} is the aligned straight dislocation density, cm^{-2}.
ρ_d^{en} is the nonaligned straight dislocation density, cm^{-2}.
Z_v^a is the vacancy-aligned dislocation bias factor.
Z_v^n is the vacancy nonaligned dislocation bias factor.

3.2.3 Vacancy emission from interstitial loops Due to the inherent geometrical curvature of an interstitial loop, the vacancy concentration at the edge of the loop differs from the bulk thermal vacancy concentration. The actual vacancy concentration is controlled by the stacking fault energy and the line tension of the loop. The various emission rates are written as:

$$P_d^{ila} = D_v C_v(r_{il}^a) Z_v^a \exp(\sigma\Omega/kT)\rho_d^{ila} \tag{20}$$

$$P_d^{iln} = D_v C_v(r_{il}^n) Z_v^n \rho_d^{iln} \tag{21}$$

$$P_d^i = P_d^{ila} + P_d^{iln} \tag{22}$$

The equilibrium vacancy concentration at the edge of an interstitial dislocation loop of radius r_{il}^a is:

$$C_v(r_{il}^a) = C_v^e \exp - \left(\frac{\{\gamma_{sf} + F_{el}(r_{il}^a)\}b^2}{kT}\right) \tag{23}$$

$$F_{el}(r_{il}^a) = \frac{\mu b^2}{(1-\nu)4\pi(r_{il}^a + b)} \ln\left(\frac{r_{il}^a + b}{b}\right) \tag{24}$$

where

P_d^{ila} is the fractional rate of vacancy emission from aligned interstitial loops, s^{-1}.
P_d^{iln} is the fractional rate of vacancy emission from nonaligned interstitial loops, s^{-1}.
P_d^{il} is the total fractional rate of vacancy emission from all interstitial loops, s^{-1}.
γ_{sf} is the stacking fault-energy, $eV\ cm^{-2}$.
$F_{el}(r_{il}^a)$ is the elastic energy of a dislocation loop of radius r_{il}^a, $eV\ cm^{-2}$.
μ is the shear modulus, $eV\ cm^{-3}$.
ν is the Poisson's ratio.

3.2.4 Vacancy emission from vacancy loops The probability of vacancy emission from a faulted vacancy dislocation loop that is parallel to the applied stress is expressed as:

$$C_v^e \exp\left\{\frac{[\gamma_{sf} + F_{el}(r_{vl})]b^2}{kT}\right\} \tag{25}$$

while for vacancy loops aligned perpendicular to the stress it is written as:

$$C_v^e \exp(\sigma\Omega/kT) \exp\left\{\frac{[\gamma_{sf} + F_{el}(r_{vl})]b^2}{kT}\right\} \tag{26}$$

From the last expressions, it is clear that these structures are thermally unstable and will dissolve quickly at high temperatures. The various emission rates are given by:

$$P_d^{vla} = D_v C_v(r_{vl}^a) Z_v^a \exp(\sigma\Omega/kT)\rho_d^{vla} \tag{27}$$

$$P_d^{vln} = D_v C_v(r_{vl}^n) Z_v^n \rho_d^{vln} \tag{28}$$

$$P_d^{vl} = P_d^{vla} + P_d^{vln} \tag{29}$$

$$r_{vl}^n = \sqrt{q_v^n/\pi b N_{vl}^n} \tag{30}$$

$$r_{vl}^a = \sqrt{q_v^a/\pi b N_{vl}^a} \tag{31}$$

$$C_v(r_{vl}) = C_v^e \exp\left\{\frac{\{\gamma_{sf} + F_{el}(r_{vl})\}b^2}{kT}\right\} \tag{32}$$

where $F_{el}(r_{vl})$ is defined in (24), and;

P_d^{vla} is the fractional rate of vacancy emission from aligned vacancy loops, s^{-1}.
P_d^{vln} is the fractional rate of vacancy emission from nonaligned vacancy loops, s^{-1}.
P_d^{vl} is the total fractional rate of vacancy emission from all vacancy loops, s^{-1}.
ρ_d^{vla} is the aligned vacancy loop line dislocations density, cm^{-2}.
ρ_d^{vln} is the nonaligned vacancy loop line dislocation density, cm^{-2}.

3.2.5 Total rate of vacancy emission The total rate of vacancy emission (P^e), in at./at./sec., is simply the sum of the previously derived rates.

$$P^e = P_C{}^e + P_d{}^e + P_{vl}^e + P_{il}^e \tag{33}$$

3.3 Single Point Defect Time Constants

In a mathematical formulation that incorporates the kinetic behavior of different irradiation produced species, there exists a wide range of time constants related to those different components. A time constant, λ, is defined here as the inverse of the time required to go through one e-folding change in a particular property, i.e., the ln (parameter) $= -\lambda t$.

The diffusion coefficient of single interstitials is orders of magnitude larger than the diffusion coefficient of single vacancies so that once they are created by irradiation, interstitials tend to diffuse quickly to different sinks and to annihilate vacancies. Since the time constants of single point defects depend on the sinks present at a particular instant, they are explicit functions of the metal's microstructure, and therefore, implicit functions of time. Their microstructural dependence can be simply expressed as:

$$\lambda_i = \lambda_i{}^d + \lambda_i{}^C \tag{34}$$

$$\lambda_v = \lambda_v{}^d + \lambda_v{}^C \tag{35}$$

$$\lambda_i{}^d = \rho_d D_i Z_i \tag{36}$$

$$\lambda_i{}^C = 4\pi N_C r_c D_i \tag{37}$$

$$\lambda_v{}^d = \rho_d D_v Z_v \tag{38}$$

$$\lambda_v{}^C = 4\pi N_C r_c D_v \tag{39}$$

where

λ_i is the total single interstitial time constant, s^{-1}.
λ_v is the total single vacancy time constant, s^{-1}.
$\lambda_i{}^d$ is the single interstitial time constant due to dislocations, s^{-1}.
$\lambda_i{}^C$ is the single interstitial time constant due to voids, s^{-1}.
$\lambda_v{}^d$ is the single vacancy time constant due to dislocations, s^{-1}.
$\lambda_v{}^C$ is the single vacancy time constant due to voids, s^{-1}.

3.4 Removal Rates

The importance of each type of sink on the dynamic behavior of point defects is reflected in the specific removal rate of the defect to that sink. Individual sink removal rates can be expressed as their relevant time constant, λ, multiplied by the temporal concentration of point defects. As indicated before, mutual recombination of point defects is a second order reaction that depends on the product of both concentrations. Collective point defect removal rates are expressed as:

$$P_{si} = \lambda_i C_i \tag{40}$$

$$P_{sv} = \lambda_v C_v \tag{41}$$

$$P_r = \alpha C_v C_i \tag{42}$$

$$\alpha = g(\nu_i \exp(-E_i{}^m/kT) + \nu_v \exp(-E_v{}^m/kT)) \tag{43}$$

where

P_{si} is the total sink removal rate for interstitials, s^{-1}.
P_{sv} is the total sink removal rate for vacancies, s^{-1}.
P_r is the total recombination rate of vacancies and interstitials, s^{-1}.
α is the recombination coefficient, s^{-1}.
g is the number of unstable sites around a vacancy.
$\nu_i \exp(-E_v{}^m/kT)$ is the interstitial jump frequency, s^{-1}.
$\nu_v \exp(-E_v{}^m/kT)$ is the vacancy jump frequency, s^{-1}.

3.5 Vacancy Loop Behavior

If b is the magnitude of the Burger's vector, the atomic volume is approximated by b^3 and n_{vl} is the fractional concentration† of vacancy loops created per second, then the number of vacancies in a vacancy loop is;

$$\pi r_{vl}^2 b/b^3 \tag{44}$$

† Here fractional concentration refers to number of loops per atomic lattice site.

and the rate at which loops are produced is

$$n_{vl} = \frac{\varepsilon P b^2}{\pi r_{vl}^2(0)} \tag{45}$$

When a vacancy loop has formed, it will immediately act as an interstitial sink because of the dislocation character of its perimeter. It will then instantly be attacked by interstitials and consequently shrink. At high temperatures the loops will also shrink by thermal emission and this process will be greatly assisted by the large line tension of such small loops and by the stacking fault energy if the loops remain faulted. Thus, each loop will have a finite lifetime τ and the number of vacancy loops per unit volume, N_{vl}, present at any time t is given by the simple rate equation

$$\frac{dN_{vl}(t)}{dt} = \frac{n_{vl}}{b^3} - \frac{N_{vl}(t)}{\tau} \tag{46}$$

where the first term on the right-hand side is the loop generation rate and the second term represents the loss term due to shrinkage.

The lifetime, τ, of an individual loop is a function of time in the sense that it depends on the state of the overall sink distribution prevailing at its instant of creation. From a Taylor series expansion of $r_{vl}(t)$ we have

$$\tau = \tau(t) \simeq -\frac{r_{vl}(0)}{(dr_{vl}/dt)|_{[r_{vl}[0]]}} \tag{47}$$

In the presence of uniaxial stress in the metal, the collapse mechanism of vacancy loops is assumed to be influenced by the stress.[3] Therefore, the following processes can be easily expressed:

$$\kappa_1^{\,n} = \tfrac{2}{3}(1-f)\frac{\varepsilon P}{\pi r_{vl}^2(0) b} \tag{48}$$

$$\kappa_1^{\,a} = \tfrac{1}{3}(1+2f)\frac{\varepsilon P}{\pi r_{vl}^2(0) b} \tag{49}$$

$$\kappa_2^{\,n} = \tfrac{2}{3}(1-f)\varepsilon P \tag{50}$$

$$\kappa_2^{\,a} = \tfrac{1}{3}(1+2f)\varepsilon P \tag{51}$$

$$\Lambda_1^{\,n} = N_{vl}^n (Z_i^{\,n} D_i C_i) - Z_v^{\,n} D_v C_v + D_v C_v^{\,e} Z_v^{\,n} \times \exp\{(\gamma_{sf} + F_{el}(r_{vl}(0)))b^2 kT\}/(r_{vl}(0) b) \tag{52}$$

$$\Lambda_1^{\,a} = N_{vl}^n (Z_i^{\,a} D_i C_i - Z_v^{\,a} D_v C_v + D_v C_v^{\,e} Z_v^{\,a} \times \exp\{(\gamma_{sf} + F_{el}(r_{vl}(0)))b^2/kT\} \exp(\sigma\Omega/kT)/(r_{vl}(0) b) \tag{53}$$

$$\Lambda_2^{\,n} = (Z_i^{\,n} D_i C_i - Z_v^{\,n} D_v C_v + Z_v^{\,n} D_v C_v^{\,e} \times \exp\{(\gamma_{sf} + F_{el}(r_{vl}^n))b^2/kT\}) \times \sqrt{4\pi q_v^{\,n} N_{vl}^n / b} \tag{54}$$

$$\Lambda_2^{\,a} = (Z_i^{\,a} D_i C_i - Z_v^{\,a} D_v C_v + Z_v^{\,a} D_v C_v^{\,e} \exp\{(\gamma_{sf} + F_{el}(r_{vl}^a))b^2/kT\} \times \exp(\sigma\Omega/kT))\sqrt{4\pi q_v^{\,a} N_{vl}^a / b} \tag{55}$$

where

- $\kappa_1^{\,n}$ is the generation rate of nonaligned vacancy loops, $cm^{-3}s^{-1}$.
- κ_{1a} is the generation rate of aligned vacancy loops, $cm^{-3}\,s^{-1}$.
- $\kappa_2^{\,n}$ is the generation rate of the fraction of vacancies tied up in nonaligned vacancy loops, s^{-1}.
- $\kappa_2^{\,a}$ is the generation rate of the fraction of vacancies tied up in aligned vacancy loops, s^{-1}.
- $\Lambda_1^{\,n}$ is the decay rate of nonaligned vacancy loops, $s^{-1}\,cm^{-3}$.
- $\Lambda_1^{\,a}$ is the decay rate of aligned vacancy loops, $s^{-1}\,cm^{-3}$.
- $\Lambda_2^{\,n}$ is the decay rate of the fraction of vacancies tied up in nonaligned vacancy loops, $s^{-1}\,cm^{-3}$.
- $\Lambda_2^{\,a}$ is the decay rate of the fraction of vacancies tied up in aligned vacancy loops, $s^{-1}\,cm^{-3}$.
- ε is the fraction of vacancies directly produced in vacancy loops (i.e., collision cascade collapse efficiency).
- $r_{vl}(0)$ is the initial radius of a vacancy loop formed in a cascade, cm.
- f is the fraction of total loop population that are aligned perpendicular to applied stress

$$f = \frac{\exp(\sigma\Omega n/kT) - 1}{\exp(\sigma\Omega n/kT) + 2} \tag{56}$$

$n \approx 10$ is the number of point defects defining a planar nucleus, as derived by Brailsford and Bullough (2).

3.6 *Swelling*

The instantaneous percent swelling can now be easily calculated as the number of voids per unit volume multiplied by the average volume of each, thus;

$$S\% = \frac{\Delta V}{V_0}\% = \tfrac{4}{3}\pi r_c^{\,3} N_C \tag{57}$$

4 FINAL STATE SPACE REPRESENTATION OF RATE EQUATIONS FOR FDRT

In state space, at a particular time, there is one and only one point that defines completely the state of the metal under irradiation. The dimensions of this space depend on the system of equations chosen to simulate the metal's response to irradiation. Based on our general treatment of Section 2, and reverting back to vector notation, the following time derivatives of 10 different components are obtained:

$$\dot{r}_c = \dot{Y}(1) = \frac{1}{Y(1)}\Big[(D_v Y(5) - D_i Y(6) - D_v C_v^e \exp\left\{\left(\frac{2\gamma}{Y(1)} - P_g(Y(1), N_g)\right)\frac{\Omega}{kT}\right\}\Big] \quad (58)$$

$$\dot{r}_{il}^n = \dot{Y}(2) = \frac{1}{b}[D_v Z_v^n Y(6) - D_v Z_v^n Y(5) + D_v Z_v^n C_v(Y(2))] \quad (59)$$

$$\dot{N}_{vl}^n = \dot{Y}(3) = \kappa_1^n - \Lambda_1^n \quad (60)$$

$$\dot{q}_v^n = \dot{Y}(4) = \kappa_2^n - \Lambda_2^n \quad (61)$$

$$\dot{C}_v = \dot{Y}(5) = (1-\varepsilon)P + P^e - P_{sv} - P_r \quad (62)$$

$$\dot{C}_i = \dot{Y}(6) = P - P_{si} - P_r \quad (63)$$

$$\dot{r}_{il}^a = \dot{Y}(7) = \frac{1}{b}[D_i Z_i^a Y(6) - D_v Z_v^a Y(5) + D_v Z_v^a C_v(Y(7)) \exp(\sigma\Omega/kT)] \quad (64)$$

$$\dot{N}_{vl}^a = \dot{Y}(8) = \kappa_1^a - \Lambda_1^a \quad (65)$$

$$\dot{q}_v^a = \dot{Y}(9) = \kappa_2^a - \Lambda_2^a \quad (66)$$

$$\dot{e} = \dot{Y}(10) = \rho_d^{ea}\{((Z_i^a - Z_i^n)D_i Y(6) + (Z_v^n - Z_v^a)D_v Y(5)) + Z_v^a D_v C_v^e \exp(\sigma\Omega/kT) - Z_v^n D_v C_v^e\} \quad (67)$$

It is this set of nonlinear, first order differential equations which form the basis for the FDRT. The major difference between this set of equations and the smaller set used by Bullough and co-workers[1–3] is the inclusion of (62) and (63) and the allowance of all the other variables to depend on the time varying values of C_v and C_i.

5 NUMERICAL SOLUTION OF RATE EQUATIONS

Equation (58) to (67) represent a system of stiff ordinary differential equations (ODE's) which are difficult to solve by standard numerical techniques. Most conventional methods for solving ODE's require incremental values of the time (t) commensurate with τ_i, while the size $|T - t_0|$ of the problem range is commensurate with max τ_i. As a result, the problem cannot be run to completion in a reasonable number of time steps.

A FORTRAN Computer Code, TRANSWELL,[7,11] has been developed to solve the previous system of equations under a variety of irradiation conditions. Among TRANSWELL's subroutines is the GEAR package[12] for the solution of the initial value problem for systems of ODE's that have the form

$$\dot{y} = f(y, t) \quad (68)$$

or more specifically,

$$\frac{dy_i(t)}{dt} = f(y_1(t), \ldots, y_N(t), t) \quad (69)$$

where y, y and f are vectors of length $N \geqslant 1$.

The basic methods used for the solution are of implicit linear multistep type. The implicitness of the basic formulae requires an algebraic system of equations be solved at each time step.

The GEAR package allows the step size and the order to vary in a dynamic way throughout the problem. The system of equations described in the previous section is a highly stiff system due to the wide range of time constants for the different components. This behavior is particularly important in transient or pulsed irradiation analysis,[11,13] as briefly described in the next section.

6 EXAMPLE OF APPLICATION TO TRANSIENT IRRADIATION

6.1 1 MeV Electron Irradiation of M316 S.S.

Since collision cascades are not produced in electron irradiated metals, vacancy loop formation is not then expected and the cascade efficiency can be set equal to zero. Furthermore, if the sample is not subject to external stresses we will have only four components of the vector **Y**; $Y(1)$, $Y(2)$, $Y(5)$ and $Y(6)$ or r_c, r_{il}^n, C_v and C_i respectively.

The parameters for M316 steel (solution treated) are those of Bullough, *et al.*[3] The calculations were performed at 600°C with an experimentally measured void concentration[14] while the interstitial loop concentration was taken at 10^{14} loops/cm^3.

The four equations for r_c, r_{il}^n, C_v and C_i are then solved numerically with the initial conditions

$$r_C(0) = 10\ \text{Å} \tag{70}$$

$$r_{il}(0) = \sqrt{4r_C^3(0)N_C/3bN_{il}} \tag{71}$$

$$P_g(0) < 2\gamma/r_C(0). \tag{72}$$

We assume here that the microstructure has already nucleated and its time dependence is studied after the radiation has begun. The time dependent behavior of the vacancy and interstitial concentrations are shown in Figure 1. At irradiation

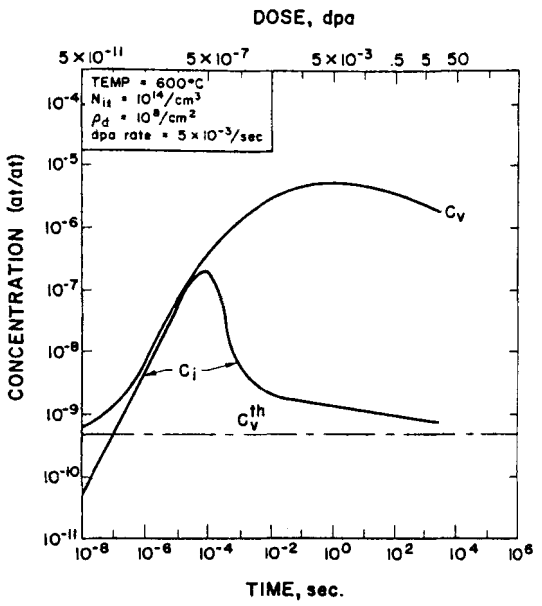

FIGURE 1 Point defect concentrations in electron irradiated M316 S.S. using the fully dynamic rate theory (FDRT).

times of the order of the first few microseconds, neither interstitials nor vacancies are mobile enough to migrate to neutral and biased sinks. Also their concentrations will be so low that the mutual recombination is negligible. Under these conditions the rate of change of the concentration of vacancies and interstitials is almost equal to the production rate. In Figure 1 the initial slope of $C_i(t)$ and $C_v(t)$ is about 5×10^{-3} at./at./sec.; the actual production rate. The build-up of the interstitial concentration coupled with their high mobility will cause the interstitial sink removal rate to increase for ~10 microseconds. The concurrent build-up of the vacancy concentration also produces a high recombination rate (which is proportional to C_vC_i). Consequently the total interstitial concentration passes through a maximum and then decreases in value as a function of time. As time progresses, the high vacancy concentration and the high mobility of vacancies will result in a vacancy sink removal rate which increases with time. After a few vacancy mean lifetimes the vacancy concentration will decrease with time producing the broad maximum as shown in Figure 1.

The absolute defect removal rates are shown in Figure 2. As expected, the interstitial sink removal is high at short times ($\sim 10^{-5}$ seconds) while the re-

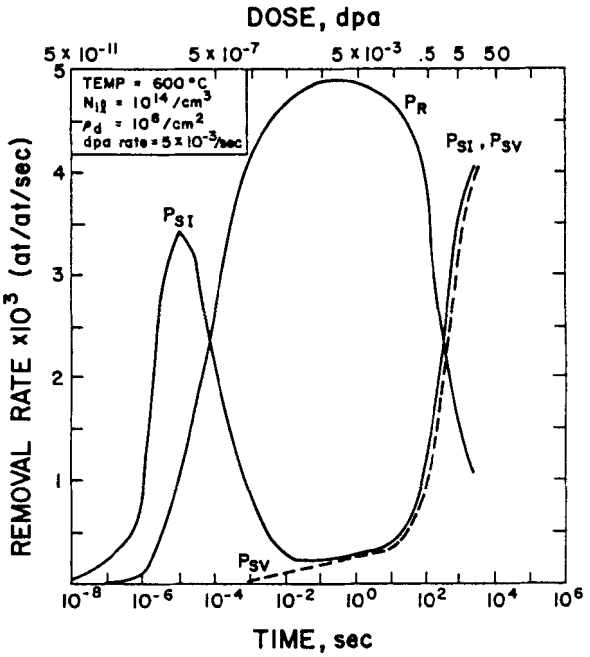

FIGURE 2 Removal rates P_R, P_{SI}, P_{SV} in electron irradiated M316 S.S. using the fully dynamic rate theory (FDRT).

combination rates are low. The build-up of vacancy concentration causes the recombination rate to dominate after $\sim 10^{-4}$ seconds. Eventually the build-up of the dislocation loop and void sinks causes the point defect removal rates to become dominant again after ~100 seconds (~0.5 dpa). Note that in Figure 2, P_R is the point defect mutual recombination rate, P_{SI} is the total sink removal rate for interstitials and P_{SV} is the total sink removal rate for vacancies in units of at./at./sec.

The significance of Figures 1 and 2 is that there is a complex time relation between point defect concentration rates and the dynamically changing microstructure. It is important to recognize the

chronology of the defect structure development, because the microstructural description at 10 or 100 dpa may be entirely different than that during the early stages of irradiation.

6.2 *Pulsed neutron simulation of 316 S.S.*

The pulsed nature of the energy release mechanism in a laser fusion reaction will produce bursts of neutrons which will cause damage in the first structural walls of a reactor for times lasting from 0.1 to 1 microsecond.[15] The instantaneous damage rates can be in the neighborhood of 10 dpa per second. In order to illustrate the flexibility of the FDRT in handling this situation we have calculated the vacancy and interstitial concentration in 316 S.S. at 600°C for one burst of neutrons and followed their behavior for 10 seconds after the damage has been produced. The results are given in Figure 3. We have used the Bullough *et al.*[3] temperature dependent void and interstitial loop concentrations, and included vacancy loop formation with $\varepsilon = 4.4\%$ and $Z_i = 1.08$. The vacancy concentration is assumed to start at the equilibrium value given in Figure 3 and we see that over the period of the pulse, the defect concentrations are a linear function of time. Immediately after the damage pulse is completed the interstitial concentration drops to essentially zero in 10 microseconds while the vacancy concentration drops only slightly due to recombination in the first few microseconds and then remains at the 10^{-6} level for ~10 milliseconds. Thereafter, the vacancies start to migrate and by 0.1 second after the damage is produced the vacancy concentration drops below that at the edge of a 30 Å diameter void. This indicates that such a void will tend to anneal once the vacancy concentration drops below that value and the void should shrink.

The reason the vacancy concentration does not return to the equilibrium value is due to thermal emission of vacancies by the dislocation microstructure and by the voids. The relationship between the microstructure and amount of annealing between damage pulses is very complex and will be examined in more detail in future papers. The main point to be illustrated by this example is that the FDRT can adequately handle the rapidly changing defect and microstructural conditions so as to describe the behavior during and after transient irradiation.

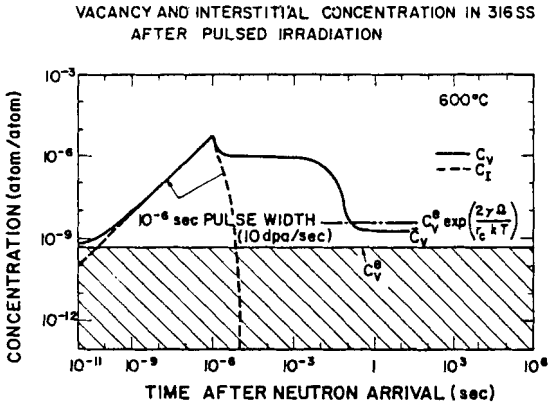

FIGURE 3 Vacancy and interstitial concentration in 316 S.S. after pulsed irradiation.

7 CONCLUSIONS

The Fully Dynamic Rate Theory (FDRT) can be used to describe rapidly changing defect concentrations and microstructural features during transient irradiation. Coupled with appropriate nucleation conditions, such description should be extremely valuable in analyzing the early stages of irradiation in a fission reactor, during heavy ion bombardment simulation studies, or in HVEM damage investigations. The FDRT should also be helpful in analyzing the transient conditions present in pulsed nuclear sources such as inertial confinement fusion reactors or rastered beam heavy ion implantation devices. The TRANSWELL Code, developed to solve the time dependent equations for the major microstructural defects, can efficiently accommodate the widely varying time frames of the above nuclear facilities.

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