

THE USE OF THE FULLY DYNAMIC RATE THEORY TO PREDICT VOID GROWTH IN METALS

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A Fully Dynamic Rate Theory (FDRT) has been used in the computer code TRANSWELL to analyze the response of metals during steady-state and pulsed irradiations. This paper correlates the FDRT with experimental data on void growth in metals. It is shown that the theory is successful in predicting the swelling behavior of different metals over a wide range of temperatures, dose rates, bombarding particles and irradiation time structures. Swelling of 316 stainless steel, Aluminum and Nickel bombarded with heavy ions, neutrons, and electrons at dose rates varying from 10^{-6} to 10^{-1} dpa/second is studied for both steady-state and pulsed irradiations.

1 INTRODUCTION

A recent analysis of the radiation pattern expected in laser fusion has revealed that the irradiation damage to the structural components will be a complex function of time.^{1,2} The response of metals to such pulsed irradiation is extremely important to the viability of this energy source and it is important to lay the theoretical base for the study of the response of metals under such transient conditions. One materials response which has been shown to degrade the performance of metals during high temperature neutron irradiation is the formation and growth of voids. The rate theory of void growth^{3,4} has been successfully applied to the high fluence steady-state irradiation case but such a model is not appropriate to handle the dynamic nature of microstructural changes taking place with time during non-steady-state irradiations. This situation has been handled by the development of the Fully Dynamic Rate Theory (FDRT) described elsewhere,⁵ and the basic features of the FDRT are incorporated in the TRANSWELL⁶ computer code. The object of this paper is to correlate the results from TRANSWELL with a wide range of steady-state and pulsed experimental data reported in the literature.

2 GENERAL APPROACH

In the FDRT formulation of the rate theory, non-linear rate equations are solved every time step. Vacancy and interstitial fractional concentrations (C_v and C_i) are described by the following equations:

$$\frac{dC_v}{dt} = (1 - \varepsilon)P + P^e - P_{sv} - P_r \quad (1)$$

$$\frac{dC_i}{dt} = P - P_{si} - P_r \quad (2)$$

where ε is the fraction of vacancies produced directly in vacancy loops (cascade efficiency), P is the point defect production rate (at/at/s), P^e is the thermal vacancy emission rate from all microstructural components (at/at/s), $P_{sv,si}$ are the vacancy/interstitial sink removal rates (at/at/s), $P_r = \alpha C_v C_i$ is the point defect bulk recombination rate (at/at/s) and α is the recombination coefficient (s^{-1}). Equations (1) and (2) are solved with four more equations for the number density of vacancy loops, the concentration of vacancies in loops, the average interstitial loop size and the average void size. The

basic work of Bullough, Eyre and Krishan⁴ has been extended to include time-dependent point defect behavior as described in detail in Ref. 5.

Examination of the literature reveals that not all the data required are available to allow a detailed comparison between theoretical models and experimental results. As a minimum, a successful growth model would require the following input: the saturation interstitial dislocation loop and void density, dpa rate, temperature, information about the displacement spike efficiency (e.g., single atom displacements by electrons *vs.* large cascade spikes from neutrons and heavy ions) and the appropriate material defect parameters. Our studies revealed that some of the above properties have been simultaneously measured in electron, heavy ion and/or neutron irradiated 316 stainless steel, Ni and Al. We have carefully chosen such data from the literature on these materials and we compare this data to the predicted results from the TRANSWELL code.

Before proceeding with the comparison, the reader should recognize several basic assumptions in the FDRT and the specific boundary conditions that were applied to this study. The two main assumptions are:

1) Nucleation of new voids and interstitial loops has ceased and the existing microstructure is merely in a state of growth (one important exception to this statement is the fact that vacancy loops are continually being formed and dissolving.⁴

2) All of the sinks are homogeneously distributed in the metal. The latter assumption is easy to justify when the defect density and dpa rates are high.³

Two specific boundary conditions for this work are:

1) The dose used in the simulation study corresponds to an actual dose measured *after* the incubation period for voids

2) To separate nucleation from growth effects, we assume that after nucleation has ceased, the irradiation is turned off, and the point defect concentrations return to thermal equilibrium. Then irradiation was turned on again to study the growth aspect of swelling.

With the above constraints, two of the main outputs from the model are the growth rates of voids and dislocation loops. The resultant swelling is easily calculated from the void density and volume.

In all the computer simulations studied we also assumed the following:

a) A simple temperature dependent nucleation relationship for the number densities of voids and interstitial loops at saturation of the form:

$$N_c^s = N_c^0 \exp \{E_c(eV)/kT\} \quad (3)$$

$$N_{ii}^s = N_{ii}^0 \exp \{E_{ii}(eV)/kT\} \quad (4)$$

where N_c^0 , N_{ii}^0 , E_c and E_{ii} are experimentally determined or when not available, computer fitted quantities.

b) Initial "free growth" void radius defined as that radius at which a void can freely grow at a certain temperature without the assistance of internal gas pressure.

c) Initial vacancy loop radius determined by the number of vacancies in a collision cascade (about 15 Å).

d) A dislocation interstitial bias factor (Z_i) adjusted to fit the metal's swelling behavior under *all* conditions.

It is also important to understand the transient behavior of the microstructure when the irradiation is turned off, then back on again. The buildup and decay of the vacancy and interstitial concentrations as well as the behavior of voids and loops between irradiation "pulses" can lend some understanding as to the ultimate response of the material. One of the crucial concepts in the application of FDRT to pulsed irradiation is that this approach allows the voids and loops to anneal out or grow in between pulses when there is no damage being produced. A more complete discussion of this effect is given in Ref. 7.

3 COMPARISON WITH EXPERIMENTS

The TRANSWELL computer code is designed primarily for dealing with metals during pulsed or transient irradiations. Unfortunately only a few pulsed experiments have been performed to date^{8,9,20} under very restricted sets of conditions. Because the problem is so complex, it is difficult to assess pulsed experiments without investigating the constituents of FDRT independently. Therefore, the basic ingredients of the theory will be evaluated by comparing the computer results with various sets of experimental data, chosen to test particular aspects of the model.

3.1 Steady-State Irradiation Experiments

We will first concentrate on the steady-state experimental data, and conclude by examining some data

TABLE I
Metal parameters used in the computer code TRANSWELL

Parameter	Symbol	Stainless steel	Nickel	Aluminum
Vacancy formation energy (eV)	E_v^f	1.60	1.39	0.70
Interstitial formation energy (eV)	E_i^f	4.00	4.08	3.20
Vacancy migration energy (eV)	E_v^m	1.30	1.38	0.57
Interstitial migration energy (eV)	E_i^m	0.20	0.15	0.10
Vacancy diffusion coefficient pre-exponential (cm ² /sec)	D_v^0	0.58	0.06	0.045
Interstitial diffusion coefficient pre-exponential (cm ² /sec)	D_i^0	0.001	0.12	0.08
Burger's vector (Å)	b	2.00	2.50	2.00
Surface energy (eV/cm ²)	γ	1.25×10^{15}	6.24×10^{14}	6.24×10^{14}
Atomic volume (Å ³)	Ω	8.00	15.63	8.00
Stacking fault energy (eV/cm ²)	γ_{sf}	9.20×10^{12}	2.496×10^{14}	1.24×10^{14}
Shear modulus (dynes/cm ²)	μ	2.83×10^{11}	9.47×10^{11}	2.65×10^{11}
Poisson's ratio	ν	0.291	0.276	0.347
Ratio of recombination to diffusion (cm ⁻²)	α/D_i	10^{16}	10^{15}	10^{16}

TABLE II
Empirical input parameters

	Stainless steel		
	Heavy Ion	Neutron	Electron
N_{ii}^0	1.34×10^{-4}	—	6.7×10^{-3}
N_c^0	3.15×10^{11}	—	6.5×10^8
E_{ii}	2.8	—	2.8
E_c	0.625	—	1.0
ε	1.2–4.4	—	0.0
Z_i	1.025–1.08	—	1.025–1.08
	Aluminum		
N_{ii}^0	7.6×10^8	10^{14} (at 55°C)	—
N_c^0	1.25×10^8	2.8×10^6	—
E_{ii}^0	0.25	0.0 (at 55°C)	—
E_c	0.55	0.55	—
ε	0.001	0.001	—
Z_i	1.015	1.015	—
	Nickel		
N_{ii}^0	10^9	—	1×10^{15} (at 450°C)
N_c^0	Experiment	—	8×10^{15} (at 450°C)
E_{ii}^0	1.0	—	0.0 (at 450°C)
E_c	Experiment	—	0.0 (at 450°C)
ε	0.01	—	0.0
Z_i	1.022	—	1.022

from pulsed studies. The fundamental material data, used in our calculations for 316 S.S., Al and Ni, is given in Table I, while the empirical input for the calculations is contained in Table II.

3.1.1 Solution treated 316 S.S.

3.1.1.1 Ion irradiation In this computer simulation, the initial void radius ($r_c(0)$) was varied

linearly between 10 and 40 Å in the temperature range 400 to 700°C. The initial interstitial loop radius ($r_{ii}(0)$) has been calculated by equating the number of vacancies in visible voids to the number of interstitials in visible interstitial loops. This procedure for calculating $r_{ii}(0)$ has been followed for all subsequent computations.

The TRANSWELL predicted swelling of 316 S.S. is compared with experiment in Figure 1. It can be

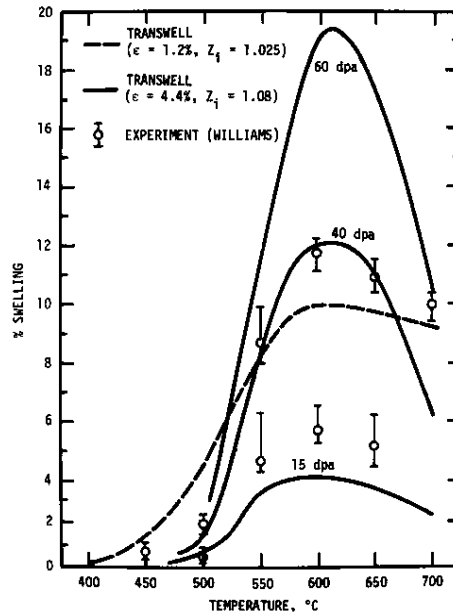


FIGURE 1 Comparison between the Fully Dynamic Rate Theory and experimental results of T. M. Williams¹⁰ for the temperature dependence of void swelling in M 316 S.S. irradiated with 22 MeV C⁺⁺ ions at a dose rate of 10^{-3} dpa/s.

seen from the figure that the correlation is not solely dependent on the *individual* values of ϵ and Z_p , but rather on the combination of these values. One notices that $Z_i = 1.025$ and $\epsilon = 1.2\%$ would give roughly the same results.

3.1.1.2. Electron irradiation Makin and Walters¹¹ studied the effect of void density on the swelling of type 316 S.S. irradiated in the HVEM and we have used their results for low (0.04%) carbon concentrations. Void and loop concentrations were fitted by the parameters of Table II.

The initial void radius was linearly varied between 10 and 40 Å in the temperature range 400 to 750°C to allow for initial void growth at higher temperatures.

The temperature dependence of the swelling of ST 316 stainless steel under electron irradiation is compared with experiment in Figure 2. The agreement in this figure is quite good but it must be emphasized that the results are sensitive to the value of Z_i used. It can be seen from the same figure that changing the bias factor from 1.08 to 1.02 would drop the swelling at 500°C and 30 dpa by a factor of 4, and at the peak temperature by a factor of 6.

3.1.2 Aluminum irradiation

3.1.2.1 Neutron irradiated aluminum Packan¹² has conducted neutron experiments on pure aluminum in HFIR irradiation environment. Void

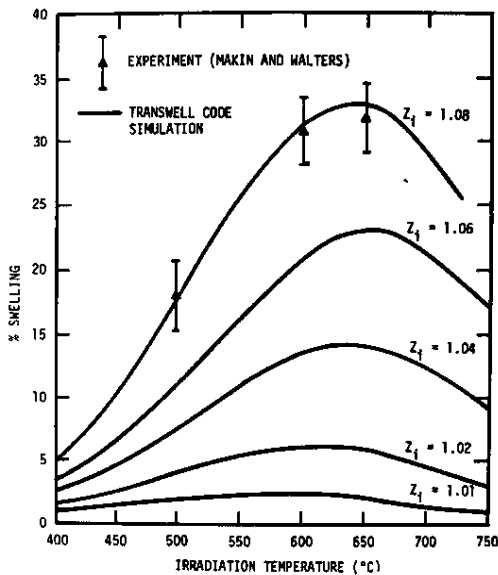


FIGURE 2 The temperature dependence of swelling in electron irradiated ST 316 S.S. at 30 dpa and a dose rate of 5×10^{-3} dpa/sec. Data from Ref. 11.

formation in high purity aluminum resulting from irradiation to fluences between 1.5×10^{19} and 1.6×10^{22} neutrons/cm² ($E > 0.1$ MeV) at a temperature of $55 \pm 5^\circ\text{C}$ was studied, primarily by means of transmission electron microscopy. Void size distribution curves were obtained for all fluences, and from these the mean void radius was found to increase in proportion to the irradiation time raised to the one-sixth power. The void concentration displayed a fluence dependence best described by a power law, $N_c \sim (\phi t)^a$, in which the exponent a decreased from 2.0 at 10^{19} neutrons/cm² down to only 0.1 at 10^{22} neutrons/cm². Treating the swelling with an analogous power relation, $\Delta V/V_0 \sim (\phi t)^b$, a similar saturation effect was observed, with the fluence exponent b decreasing roughly from 5/2 to 1/2 over the range of fluence studied.

Calculations of displacements per atom were made using HFIR spectrum.¹³ The displacement cross sections calculated with a threshold displacement energy of 16 eV for Al are reported elsewhere.¹⁴ A value of 1.3 dpa was found to correspond to a fluence of 10^{21} n/cm²/s for neutrons with energy greater than 0.1 MeV.

The displacement rate was found to vary between 1×10^{-6} and 1.6×10^{-6} dpa/sec. Therefore, an average value of 1.3×10^{-6} dpa/sec was used in the calculations. Other experiments on neutron irradiated aluminum^{15,16} indicated that the saturation void number density can be fitted to the expression of Table II. A gas production rate of 3×10^{-12} at/at/s was assumed in these calculations.

Figure 3 shows the agreement between Packan's experimental swelling results and TRANSWELL calculations. The same figure shows the agreement between measured average void sizes and computer

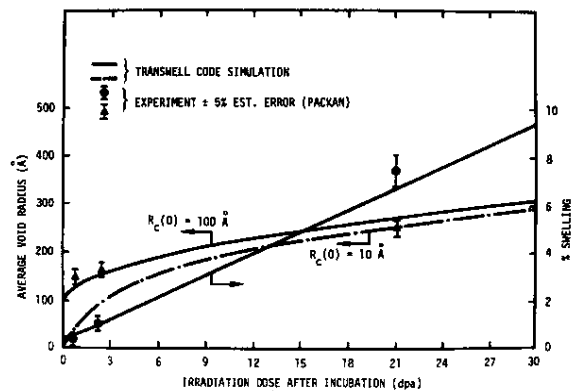


FIGURE 3 Comparison between measured and calculated values of both swelling and mean void radius of aluminum under neutron irradiation. Data from Ref. 12.

calculated values if one starts with void sizes of 10 or 100 Å radius. The agreement is shown to be still good even if the starting void radius is very small, indicating that the somewhat arbitrary choice of a free growth radius is not too restrictive in the model.

3.1.2.2 Ion irradiated aluminum Irradiation of aluminum with 400 keV Al ions was carried out using the Harwell 500 kV Cockroft-Walton accelerator by Mazey, Bullough, and Brailsford.¹⁷ The temperature dependence of void swelling was investigated in non-helium doped 1100 grade aluminum by 400 keV Al⁺ irradiation to 1×10^{17} ions/cm² at temperatures of 75, 100, 150, 200 and 250°C.

Material and empirical input parameters of tables I and II have been used with a displacement rate of 10^{-3} dpa/s to simulate Mazey's experiment up to a total accumulated dose of 104 dpa. The initial void radius was held constant at 40 Å.

A comparison of TRANSWELL results to the experimentally determined swelling is shown in Figure 4. The results of the calculations seem to indicate that aluminum is not as efficient as steel in retaining vacancies in collision cascades (e.g., $\epsilon = 0.001$ in Al vs. 0.012–0.044 for 316 S.S.).

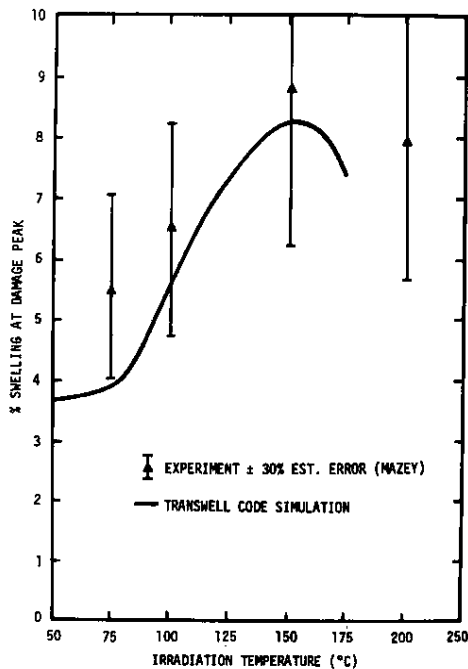


FIGURE 4 Temperature dependence of void swelling in aluminum irradiated with 400 KeV Al⁺ ions after a total dose of 104 dpa.

3.1.3 Nickel irradiation

3.1.3.1 Ion irradiated nickel The temperature dependence of void and dislocation loop structures was studied by Sprague, *et al.*⁸ in high-purity nickel irradiated with 2.8 MeV ⁵⁸Ni⁺ ions to a displacement dose of 13 displacements per atom (dpa) at a displacement rate of 7×10^{-2} dpa/sec over the temperature range to 625°C. Dislocation loops with no significant concentrations of voids were observed in specimens irradiated at 475°C and below. Specimens irradiated between 525 and 725°C contained both voids and dislocations. The maximum swelling was measured as 1.2% at 625°C.

The metal parameters of Tables I and II were adopted and the experimentally measured void densities were used in all the computer calculations.

As shown in Figure 5 there is good agreement between calculated and experimentally measured swelling values below the peak temperatures. Curve (2) shows the better agreement at high temperatures if the starting void radius was taken to be 200 Å. It was observed from all the calculations that small voids find it difficult to grow in ion or neutron irradiations without the assistance of gas for

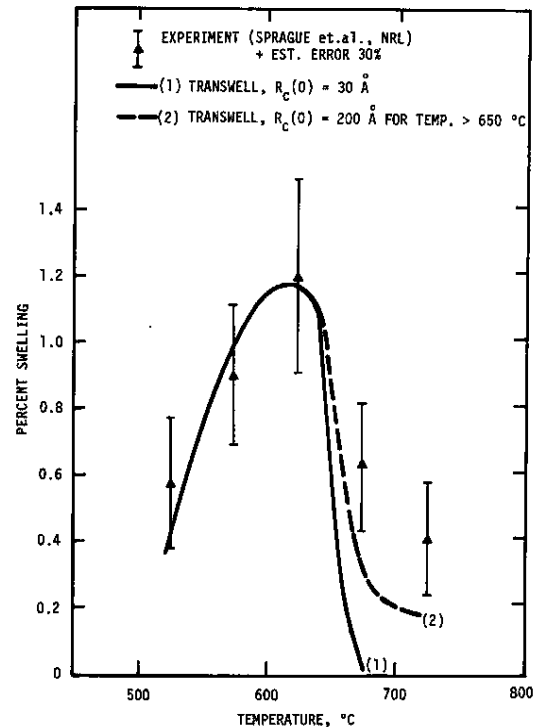


FIGURE 5 Temperature dependence of void swelling in nickel irradiated with nickel ions. The dose rate is 7×10^{-2} dpa/sec and the total dose is 13 dpa. Data from Ref. 8.

temperatures above the peak temperature. This suggested that the present growth theory can be better correlated with data at high temperatures if one starts with a "free-growth radius" as defined before which is obviously temperature as well as irradiation and microstructure dependent.

The important point to note from Figure 5 is that TRANSWELL comes remarkably close to predicting the swelling in ion bombarded Ni from 500°C to 750°C under relatively high displacement rates (7×10^{-2} dpa/sec).

3.1.3.2 Electron irradiated nickel Voids in nickel, electron irradiated at 450°C, were studied by Norris.^{18,19} The specimen was previously bombarded with 6×10^{17} He⁺ ions/m² at 50 keV and then annealed at 640°C to remove the vacancy clusters. A calculated displacement rate of 2×10^{-3} dpa/sec with an error of 20% was used. Figure 9 shows the results of this experiment with dose uncertainty of 20% and swelling uncertainty of 30%. Swelling saturation at high doses is suspected to be due to surface effects in thin foils.

Unfortunately, only total void induced swelling was reported in the electron irradiation studies of Ni, no void density, size or interstitial loop information was given. Therefore, void and loop number densities of steel at 450°C have been used, even though we realize that there may be some significant differences.

The input from Tables I and II are again used. It is found that computer simulations produce reasonable agreement with experimentally measured swelling values as shown in Figure 6. At large doses, foil surface effects impose artificial saturation limits on the swelling data, and therefore accurate comparison with the present model is difficult in that region.

3.2 Pulsed Irradiation Experiments

A recent experimental study by Potter *et al.*²⁰ has illustrated that pulsed irradiation of Ni to the same damage level and at the same damage rate as for a corresponding sample irradiated in a steady-state fashion, can give significantly different microstructural results. First, several Ni specimens were pre-injected with 5 appm of Helium and then irradiated at 600°C to 5 dpa with a steady-state damage rate of 3×10^{-3} dpa/sec (A). Three specimens were then given a further irradiation at 500°C to 5 dpa with a damage rate of 3×10^{-3} dpa/sec (B). Another three specimens were irradiated to 5 dpa in 6.75 sec pulses separated by 6.75 sec intervals (C). A total of 247

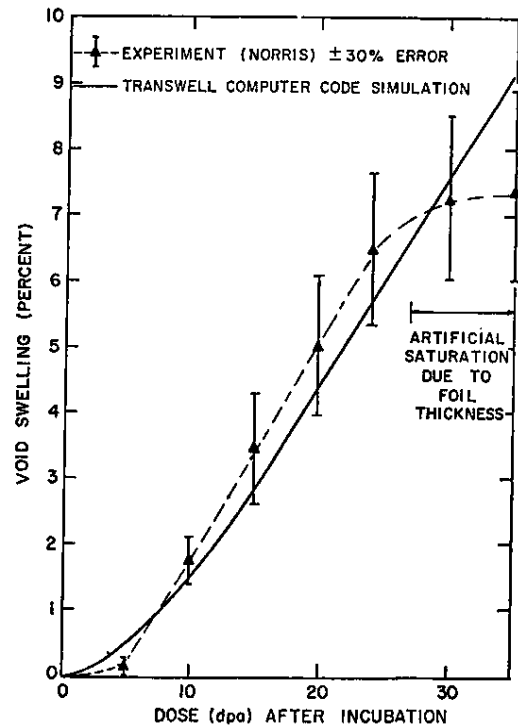


FIGURE 6 Dose dependence of void swelling in nickel irradiated with electrons at 450°C at a dose rate of 2×10^{-3} dpa/sec. Data from Ref. 18.

pulses accumulated at the end of the experiment. The resultant void structures for these irradiations are listed in Table III.

The subsequent irradiations (cases B and C) of the pre-voided samples caused the void radius to grow; from 66 to 96 Å in the steady-state irradiation and from 66 to 121 Å in the pulsed study. The void densities decreased in both of the subsequent irradiations. Even when this is taken into account, it was observed that the total incremental swelling in the pulsed study was over twice that from the steady-state irradiation.

The TRANSWELL code was then used to simulate the conditions outlined in Table III. The variation in void number density was accounted for by adjusting the value of the void density downwards in a linear fashion every one dpa of accumulated damage. First, the sink density due to vacancy loops in the pulsed irradiation experiment never reaches such a high level as in the steady-state situation because of annealing between pulses. And second, vacancy loops emit large numbers of vacancies during the annealing period, which can then end in voids without recombination with interstitials. In this beam-off time, vacancy concentration drops slowly

TABLE III

Summary of experimental void data on heavy ion irradiated Ni pulsed vs. Steady-state damage (Potter *et al.*²⁰)

Study	Temp. (°C)	Total dose (dpa)	Duration of damage pulse (sec)	Time between pulses (sec)	Number of pulses	Void radius (Å)	Void density $\times 10^{-15}$ (cm ⁻³)	$\frac{\Delta V}{V}$ %	Remarks
A	600	5 ^a	1667.00	0.00	1	66	6.3	0.84	Prevoided Subsequent irradiation of samples from A
B	500	10 ^b	1667.00	0.00	1	96	3.3	1.4	
C	500	10 ^b	6.75	6.75	247	121	2.3	2.1	

^a Contains 5 appm He.^b Includes 5 dpa from study A.

to its equilibrium value (which is $>C_v^e$ due to vacancy emission), while the interstitial concentration drops immediately ($\sim 10^{-6}$ seconds) to insignificant levels ($\sim 2.5 \times 10^{-27}$ at/at at 500°C). The slight increase in the equilibrium vacancy flux[†] (Figure 8) is due to the higher emission of vacancy loops as they shrink. The main parameter of interest, the average void radius, is plotted in Figure 7. Agreement between the void radius predicted by the model and that measured experimentally is good; within 1% for the steady-state irradiation and 10% for the pulsed irradiation. Aside from the absolute numbers, the TRANSWELL Code correctly predicted that the void swelling during this particular pulsed experiment is substantially greater than that observed during steady-state studies.

[†] The term "flux" is used for the product of the point defect diffusion coefficient and its fractional concentration (cm²/s).

The enhanced swelling in the pulsed case has been found to be due to the behavior of vacancy loops.

A more quantitative description of this effect is shown in Figure 8, where the flux of vacancies and interstitials to the voids is given along with the emission of vacancies from void surface during the 100th pulse (0.9943–2.0012 dpa). During the damage pulse, the void grows because the vacancy flux is greater than the combined interstitial and emission fluxes. When the damage pulse is over, $D_i C_i$ drops quickly to 3.16×10^{-29} cm²/sec, while the emission flux is essentially constant at 1.26×10^{-18} cm²/sec. The $D_v C_v$ term drops off with a longer life-time than $D_i C_i$, and for the entire 6.75 sec down period, the vacancy flux into the void is greater than the vacancy emission, hence the void continues to grow.

Analysis of the vacancy emission and absorption rates at the end of the annealing period for the 100th

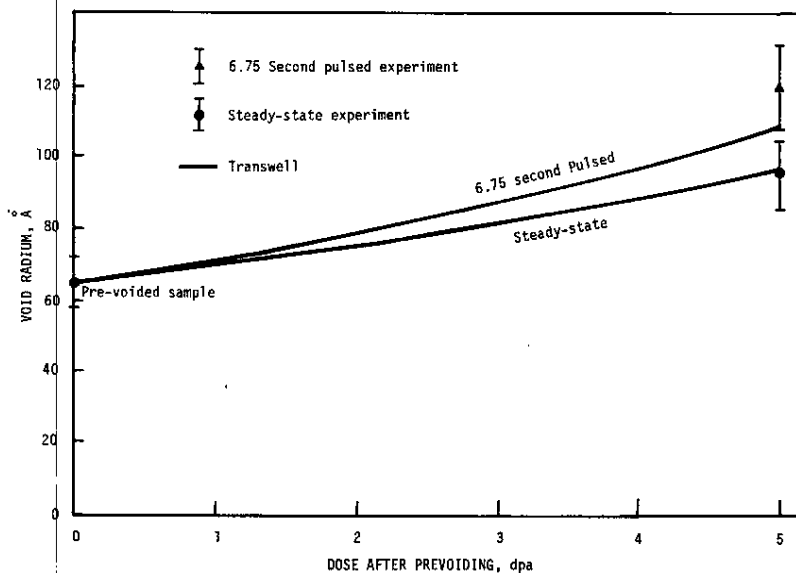


FIGURE 7 Dose dependence of steady state and pulsed average void radii in self-ion irradiated pure nickel at 500°C. Total dose is 5 dpa, dose rate = 3×10^{-3} dpa/sec and $t_{on} = t_{off} = 6.67$ sec. Data from Ref. 20.

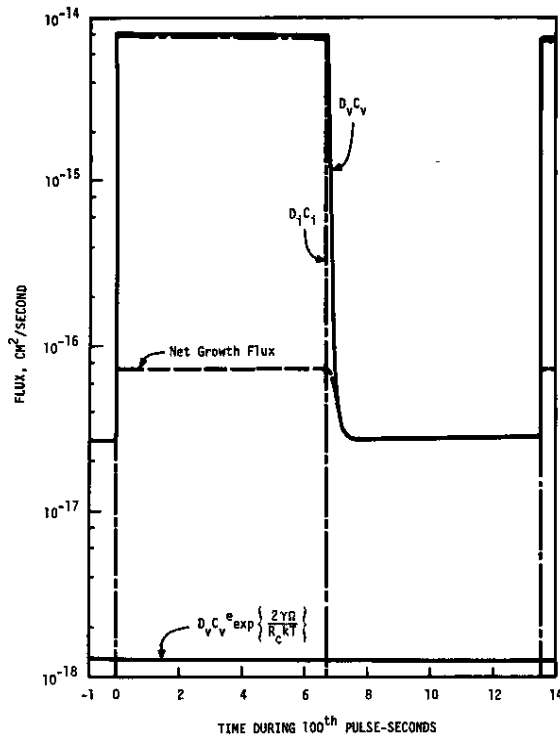


FIGURE 8 Point defect fluxes near an average void surface during the 100th pulse (0.9943–2.0012 dpa).

pulse of study C indicates that vacancy emission from vacancy loops is the cause of void growth in the interpulse period. The vacancy emission rate from vacancy loops is 9.2×10^{-6} at/at/s while their vacancy absorption rate is 5.9×10^{-6} at/at/s. The excess amount of vacancies are essentially absorbed at voids and interstitial loops. While voids emit vacancies at the rate of 6.4×10^{-9} at/at/s and interstitial loops at 4.8×10^{-10} at/at/s, vacancies are absorbed at voids with the rate of 1.4×10^{-6} at/at/s and at loops with 1.9×10^{-6} at/at/s. It is this *delayed* vacancy emission in the absence of free interstitials which accounts for the enhanced growth rate over the steady-state irradiation.

4 DISCUSSION AND GENERAL REMARKS

The fundamental reason for the growth of three dimensional cavities (voids) in irradiated materials is believed now to be understood^{3,4} and many of the contributory physical mechanisms have been identified. By adopting a more systematic approach and relating theory to experiments, as attempted in this paper, the dominant mechanisms affecting the void swelling problem can be qualitatively parameterized.

The results of the present comparison between the theory and experimental data have shown that the FDRT can predict the high temperature void swelling behavior of both pure metals and alloys. The fact that the theory predicts the temperature dependence of swelling over a wide range of displacement rates ($\sim 10^{-6}$ to $\sim 10^{-1}$ dpa/sec) and with different bombarding species (neutrons, heavy ions and electrons) is satisfying. The agreement with pulsed experiments is also gratifying. However, the reader must remember that this FDRT only applies to the situation when a quasi-steady-state void and interstitial loop density has already been established, i.e., it does not apply to the initial stages of irradiation when nucleation is still dominant.

At high temperatures ($>0.4-0.45 T/T_m$), it is noted that the theory predicts lower swelling than observed. This difficulty arises because the initial void size has to be relatively large to withstand the emission of vacancies (which is roughly proportional to $1/r$). One way to remedy this would be to use an empirically determined initial void size which is an increasing function of temperature.

The use of different spike collapse efficiencies (1.2–4.4% for steel, 0.1% for Al and 1% for Ni) to achieve agreement between theory and experiment indicates that the displacement cascade in Al is rather diffuse and inefficient at retaining vacancies. It also demonstrates that future research is required to understand the underlying phenomena that control the collision cascade collapse mechanism.

It will also be noticed from the comparison below that slightly different values of the bias factor of dislocations for interstitials were required for good agreement between the theory and experimental data.

Material	Bias Factor
316 S.S.	1.025–1.08
Al	1.015
Ni	1.022

However, one would expect that since the bias factor depends on the interaction between point defects and dislocations, which is material dependent. Therefore, different values of Z_i are assigned to different materials.

The results of this study show that a fully dynamic rate theory can accurately predict the post incubation growth behavior of voids and dislocations loops as a function of temperature in a wide variety of materials irradiated over a factor of 10^5 in

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steady-state displacement rates with widely different types of irradiation as well as with pulsed irradiation. The difficulty in finding experimental data to correlate with theory demonstrates the need for more specifically designed calibration experiments. This subject has been discussed elsewhere⁷ and specific recommendations have been made. Once such experiments are performed providing dislocation loop, and void data as a function of dose and temperature, the FDRT can be further refined and calibrated to predict the growth of voids in metals. Correlation of the theory to more extensive pulsed experiments can be accomplished by the careful choice of pulsing schemes in those experiments. Meanwhile, the examination of the time dependent microstructure predicted by FDRT is providing valuable insight to the factors promoting the deleterious void growth in irradiated metals.

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