Presented at Fusion Reactor Materials
Conference, Miami Beach, January 29–31, 1979

CONF-790125--51

SWELLING WITH INHOMOGENEOUS POINT DEFECT PRODUCTION — A CASCADE DIFFUSION THEORY*

L. K. MANSUR and W. A. COGHLAN
Oak Ridge National Laboratory, Oak Ridge, Tennessee 37830, USA

and

A. D. BRAILSFORD
Ford Motor Company, Dearborn, Michigan 48121, USA

ABSTRACT

A theoretical method is described for evaluating the effects of spatially and temporally discrete production in collision cascades on point defect concentrations and swelling in materials during irradiation. The concentrations of vacancies and interstitials at a point which result from their diffusion from all cascades in the material are calculated. Large fluctuations occur with time in the vacancy concentration. The interstitial concentration is nearly always zero except for extremely large spikes of very short duration, corresponding to the occurrence of a cascade anywhere within the sphere beyond which all generated defects are absorbed by sinks before reaching the reference point. The growth rate of a void in this cascade diffusion theory is compared to that given by the more approximate rate theory. The difference is small but increases rapidly at high temperature. Implications of this work for void nucleation, irradiation creep, and analysis of pulsed irradiations are mentioned.

*Research sponsored by the Division of Materials Sciences, U. S. Department of Energy under contract W-7405-eng-26 with the Union Carbide Corporation.

1. INTRODUCTION

The rate theory of swelling has been developed primarily for application to steady irradiations. Fusion reactors, accelerator-driven neutron sources and to a different degree fission reactors often are designed or operated on an interrupted or pulsed schedule. The radiation on-time may range from nanoseconds to more than thousands of seconds, with the off-time spanning fewer orders of magnitude. Several studies have investigated the point defect concentrations, void nucleation, and swelling behavior to be expected during pulsed irradiations on the basis of rate theory [1–6]. In general the predicted swelling is found to depend on the pulse duration and pulse interval, with some regimes of cycle times leading to small or negligible swelling and others giving results similar to steady irradiations.

One of the implicit assumptions of rate theory is that point defect generation and loss can be treated as if occurring continuously in time and at every point in the medium. Point defect generation by radiation, which occurs in cascades caused by spatially and temporally random collisions of the bombarding particles with matrix atoms. It is modeled, however, as if generated homogeneously and continuously by an infinite number of infinitesimal regions in a continuum. Pulsed irradiations have been modeled by suppressing and reactivating this continuum source.

However, it is known by considering the physical nature of point defect production [7,8] that point defect concentrations fluctuate locally even during steady irradiations. We, therefore, view the question of the effects of radiation pulsing as really a question of what *additional* effects are introduced by radiation pulsing when this is superimposed on the already locally fluctuating point defect concentrations. Yet the implications of locally fluctuating point defect concentrations are not fully understood. We therefore undertook the work to be reported in the present paper.

2. THEORY

A cascade occurs at time t_c and position ρ_c. The initial point defect concentration is modeled as a delta function $\delta(\rho-\rho_c)\ \delta(t-t_c)$. As will be shown, this is equivalent to introducing an initially localized Gaussian distribution of point defects of finite spatial extent at $t_c + \varepsilon$ where ε is a small time increment. The diffusion of point defects from such a unit cascade obeys the continuity equation

$$D_\alpha\ \nabla^2\tilde{c}_\alpha - D_\alpha S_\alpha \tilde{c}_\alpha + \delta(\rho-\rho_c)\ \delta(t-t_c) = \frac{\partial\tilde{c}_\alpha}{\partial t}\ . \tag{1}$$

The subscript α denotes either vacancies or interstitials. $D_\alpha = D^\circ_\alpha \exp(-E^m_\alpha/kT)$ is the diffusion coefficient where D°_α is a constant, E^m_α is the migration energy, k is Boltzmann's constant and T is absolute temperature. S_α is the sum of the sink strengths for point defects of all sinks and is the same as that used in rate theory. In the calculations described later, $D^\circ_v = 0.014$ cm^2/s, $E^m_i = 0.15$ eV, $E^m_v = 1.4$ eV, $S_i = 1.1 \times 10^{11}$ cm^{-2} and $S_v = 1 \times 10^{11}$ cm^{-2}. $\tilde{c}_\alpha$ is the concentration per unit volume of point defects. The overscore character denotes the explicit variability of this concentration with position and time due to cascade diffusion to distinguish the symbol from the C used in rate theory [8]. In addition the overscore character is a reminder that while the C_v in the rate theory in our convention [8] contains both the radiation produced vacancy concentration as well as the thermal vacancy concentration produced by radiation. Thus we have used continuum rate theory in accounting for diffusion and for absorption at distributed sinks. Possible corrections due to the rapid spatial and temporal variations, as they influence these processes, have not been considered. Our main interest here lies in the *direct* influence of the discrete point defect production, for which we take Eq. (1) to be adequate [10].

The unit solution to Eq. (1) is [9]

$$\tilde{c}_\alpha(\rho|\rho_c, t-t_c) = [4\pi D_\alpha (t-t_c)]^{-3/2}$$

$$\times \exp[-(\rho-\rho_c)^2/(4D_\alpha (t-t_c))] \exp[-D_\alpha S_\alpha (t-t_c)] \quad . \qquad (2)$$

The product of terms describes broadening of the cascade by diffusion and collapse of the cascade by absorption of point defects at sinks in the medium. When $(t-t_c) = (D_\alpha S_\alpha)^{-1}$, the concentration in the cascade integrated over all space decreases by e^{-1}. Thus we refer to $S_\alpha^{-1/2}$ as the absorption mean free path of the defects α. For a cascade producing ν net defects, Eq. (2) is multiplied by ν. By choosing ν it is thus possible to account for recombination within the cascade reducing the net number of defects available for diffusion.

Having the Green's function for each cascade, the next part of the problem is to develop a scheme for introducing the many cascades into the medium in a physically realistic way and then to keep track of the evolution of all cascades. This will allow the calculation of the concentration at an arbitrary reference point arising from the superposition of the time dependent contribution from every cascade.

About the reference point we construct a series of concentric spherical shells of uniform thickenss, the i'th shell having inner radius ρ_{i-1} and outer radius ρ_i. The probability of ℓ cascades in each shell of volume $V_i = (4/3)\pi(\rho_i^3-\rho_{i-1}^3)$ in a unit time is

$$P(\ell) = \exp(-\Sigma \phi V_i) (\Sigma \phi V_i)^\ell/\ell! \qquad (3)$$

where the mean number of cascades per unit time per unit volume is $\Sigma \phi$. Here $\Sigma = \sigma N$ is the macroscopic cross section for collisions producing a cascade, where σ is the microscopic cross-section, N the volume concentration of atoms, and ϕ the path length of bombarding particles per unit volume, the "flux." In the calculations illustrated later the mean number of cascades per unit time in a shell of volume V_i, $\Sigma \phi V_i$, determines the interval between cascades in a given shell,

$$\tau_i = (\Sigma \phi V_i)^{-1} \quad . \tag{4}$$

To simplify the calculation, the diffusion from all cascades occurring in shell i is initiated on an imaginary sphere whose radius is $(r_i + r_{i-1})/2$. The error of this approximation $\rightarrow 0$ as $(r_i - r_{i-1}) \rightarrow 0$. The range of shell thicknesses used is 0.02–0.14 of $S^{-1/2}$ (i.e., one to two orders of magnitude smaller than the absorption mean free path).

An efficient numerical technique has been developed to integrate the contributions from all cascades by inserting each cascade at the correct instant and position and subsequently tracking the diffusion of the point defects until the cascade has decayed completely. To specify its complete decay we refer to the rate theory concentration, C_α. Simplified quasi-steady state rate equations are sufficient

$$G_\alpha - RC_vC_i - K_\alpha C_\alpha = 0 \quad . \tag{5}$$

G_α is the generation rate per unit volume of defects of type α and, for vacancies, includes both thermal vacancy generation and radiation-induced generation as follows

$$G_v = \nu_v \Sigma \phi + \sum_J K_v^J C_v^{eJ} \quad . \tag{6}$$

Here ν_v is the net number of vacancies per cascade, $K_v^J = D_v S_v^J$ is the loss rate at sinks of type J ($\sum_J K_v^J \equiv K_v = S_v D_v$) and C_v^{eJ} is the thermal vacancy concentration at sinks of type J. In Eq. (5) C_α is the quasi-steady-state volume averaged concentration of defects of type α per unit volume. From Eqs. (5) and (6) it can be seen that C_v contains both irradiation-induced and therma concentrations of vacancies. Thermal vacancy emission is also properly accounted for in the cascade diffusion theory but by separate means as discussed below. The cascade diffusion theory concentrations therefore must be compared to

$$C_\alpha^\circ = \frac{\nu_\alpha \Sigma \phi}{D_\alpha S_\alpha} \quad . \qquad (7)$$

The superscript ° denotes the radiation-induced concentration. Equation (7) is obtained from Eq. (5) by neglecting the simple bulk recombination approximation, the second term of Eq. (5). This is to enable a direct comparison with the cascade diffusion theory, where it has not been attempted to account for the extremely complex spatial variation of recombination which results from the ability to resolve all individual cascades. The use of high sink strengths, $S \sim 10^{11}$, in the subsequent calculations minimizes the effects of recombination.

To gauge when a cascade has dissipated we apply the condition

$$\tilde{c}_\alpha < \varepsilon_c \, C_\alpha^\circ \qquad (8)$$

where ε_c is a fraction << 1. In our calculations, removing a cascade from the system when $\tilde{c}_\alpha < 10^{-6} \, C_\alpha^\circ$ at the point of observation introduces negligible error. However, before the cascade is removed a second condition must be fulfilled. It arises from the fact that just after a cascade has occured, Eq. (8) will always hold at any reference point which is a finite distance removed, until a sufficient time passes for the wave of concentration to reach the point. The cascade must not be discarded unless condition (8) holds *and* the concentration is decreasing. Thus we require also that

$$\frac{\partial \tilde{c}_\alpha(\rho_o|\rho_c,\ t-t_c)}{\partial(t - t_c)} < 0 \tag{9}$$

where ρ_o is the coordinate of the reference point, which is satisfied when

$$D_\alpha\ (t-t_c) > \frac{[9 + 4S_\alpha\ (\rho_o-\rho_c)^2]^{1/2}}{4S_\alpha} - 3\ . \tag{10}$$

Since $D_i >> D_v$, vacancy dissipation determines the time beyond which a cascade can be discarded. In typical calculations the progress of $\sim 10^3$ different cascades must be followed since these are contributing to the concentration at the reference point. After a steady state has been achieved, on the average when one cascade dissipates, another occurs by a collision at some other random position in the material. The total radiation-induced concentration at a point of observation $\tilde{C}_\alpha$ is the sum of the contributions from all cascades,

$$\tilde{C}_\alpha = \sum_m \tilde{c}_{\alpha,m}\ , \tag{11}$$

where $\tilde{c}_{\alpha,m}$ is the contribution from a cascade labeled m, where m ranges over all cascades making contributions to $\tilde{C}_\alpha$ which satisfy conditions (8) and (10).

The growth rate of a void whose environment is approximated as given by Eq. (11) may be calculated according to

$$\frac{dr_v}{dt} = \frac{\Omega}{r_V} \left\{Z_v^V D_v [\tilde{C}_v - C_v^e\ [\exp((2\gamma/r_V - P_g)\Omega/kT) - \sum_J K_v^V C_v^{eJ}/C_v^e \sum_J K_v^J] - Z_i^V D_i \tilde{c}_i\right\} \tag{12}$$

The corresponding expression using the rate theory concentrations is

$$\frac{dr_v}{dt} = \frac{\Omega}{r_v} \{Z_v^V D_v [C_v - C_v^e \exp((2\gamma/r_v - p_g)\Omega/kT - Z_i^V D_i C_i\} . \quad (13)$$

Here C_v^e is the bulk thermal vacancy concentration, Z_α^V is the capture efficiency of a void for defect α, γ is the surface tension, p_g is the gas pressure and Ω is atomic volume.

3. RESULTS

Figure 1 shows the average vacancy concentration as a function of the radius of the spherical volume of material about the reference point within which cascades are allowed to occur. The volume of material within a distance of $\sim 7S^{-1/2}$ gives an average concentration equal to the bulk averaged rate theory result. The volume within $\sim 3S^{-1/2}$ contributes $\sim 80\%$ of the average rate theory concentration. This clearly illustrates the fact that cascades occurring at large distances ($\gtrsim 7S^{-1/2}$) do not contribute to the concentration at the point of observation because of absorptive losses in the medium. The other important observation based on Fig. 1 is that the cascade diffusion theory developed here gives the same average result as the rate theory equations for C_v and C_i. This provides an independent confirmation of the validity of the rate equations for C_v and C_i for the case where recombination is unimportant. It must be stressed, however, that this alone does not establish the validity of the rate theory of void nucleation, void growth and irradiation creep, for reasons which will become apparent.

Figures 2 and 3 present the vacancy and interstitial concentrations as functions of time. The remarkable feature is the extreme fluctuation in point defect levels. The vacancy concentration is composed of a mildly fluctuating background supplied by the many more distant cascades, punctuated by abrupt spikes from the relatively few nearby cascades which raise the instantaneous concentration more than an order of magnitude. The spikes in the interstitial concentration are more pronounced and there is no background level. In Fig. 3 the time spans a random interval covering the occurrence of three cascades. The cascades occur at random distances from the reference point, which are indicated on the top of Fig. 3. The concentration rises and decays in fractions of a microsecond while these cascades are separated by more than milliseconds. In these calculations it is found that there is virtually never more than one cascade contributing to the interstitial concentration. More than this, for the vast majority of time there is zero interstitial concentration at any arbitrary point of observation.

Figure 4 compares the growth of a void, arbitrarily chosen to be of 10 nm radius, which results from the ratcheting in response to the concentrations shown in Figs. 2 and 3, using Eq. (12), with the growth resulting from averaging the instantaneous concentrations first and then computing the growth rate of the void. This latter is the rate theory approximation and uses Eq. (13). The difference with rate theory is small. However, it increases rapidly with increasing temperature. This is due in part to the fact that the thermal emission rate of vacancies from the void is treated more accurately in the cascade diffusion theory.* In the cascade-resolved case the void undergoes successive growth and shrinkage by small amounts in response to the delivery of point defects from cascades. However, the void emits thermal vacancies

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*It is also due in part to the fact that $\Delta(\Delta r)$, the differences in the increase in void radius by growth as calculated by rate theory and cascade diffusion theory, remains finite as Δr approaches zero at the temperature where thermal vacancy emission equals the net vacancy influx.

at different rates depending on its size, since the thermal equilibrium vacancy concentration at a void of radius r_v is given by

$$C_v^e(r_v) = C_v^e \exp[(2\gamma/r_v - p_g)\Omega/kT] \ . \qquad (14)$$

Since by Figs. 2 and 3 the void is usually growing in response to a (albeit rapidly varying) vacancy flux with only occasional high fluxes of interstitials it will be larger than the average of its initial and final sizes for most of the growth period. Therefore the integrated thermal emission rate of vacancies over time will be smaller than that corresponding to its average size, so that the cascade diffusion theory leads to slightly faster growth rates than the rate theory.

4. SUMMARY AND DISCUSSION

A theoretical method has been developed to account for the physically discrete generation of point defects in cascades. Large local fluctuations in vacancy and interstitial concentrations occur during a steady irradiation. The results confirm the validity of the usual bulk-averaged rate theory equations for the average vacancy and interstitial concentrations where recombination is unimportant. There are however differences in the predicted void growth using the more accurate cascade resolved diffusion theory as compared with the rate theory.

Further applications of the cascade diffusion theory can be suggested, covering virtually all areas now studied using rate theory. These include void and loop nucleation, irradiation creep, and solute segregation. The effects on all these processes of pulsed irradiations could be studied using the present cascade diffusion theory. Present nucleation theory operates with a picture of uniform concentrations of point defects leading to a statisitcal distribution of cluster sizes which are generally accumulated by random absorption or emission of single point defects. Here, however, it is found that a void embryo is subject to an extremely variable but uninterrupted vacancy flux and only infrequently to an interstitial flux. The interstitial flux comes in short bursts of high intensity. Clearly there will be some effect on the current picture of void nucleation. The implications are also significant for interstitial loop nucleation. Figure 3 suggests that interstitial loop nucleation may be possible mainly as a result of self-agglomeration of interstitials from a single cascade. Overlap of interstitials from more than one cascade is unlikely. Impurity trapping, however, offers a solution to this dilemma, since this can decrease the effective diffusion coefficient of interstitials [8]. These results may also have implications for irradiation creep by climb-enabled glide. The critical function of climb is to overcome obstacles to the glide of dislocation segments, which results in creep. The results above lead to the expectation that it is not necessary to have an overall net absorption of one or the other type point defect to obtain climb-enabled glide if a cascade occurs in the vicinity of the obstacle. Figures 2 and 3 imply that local climb can take place where an unbalanced net vacancy or interstitial flux impinges on a dislocation segment over short time increments.

5. REFERENCES

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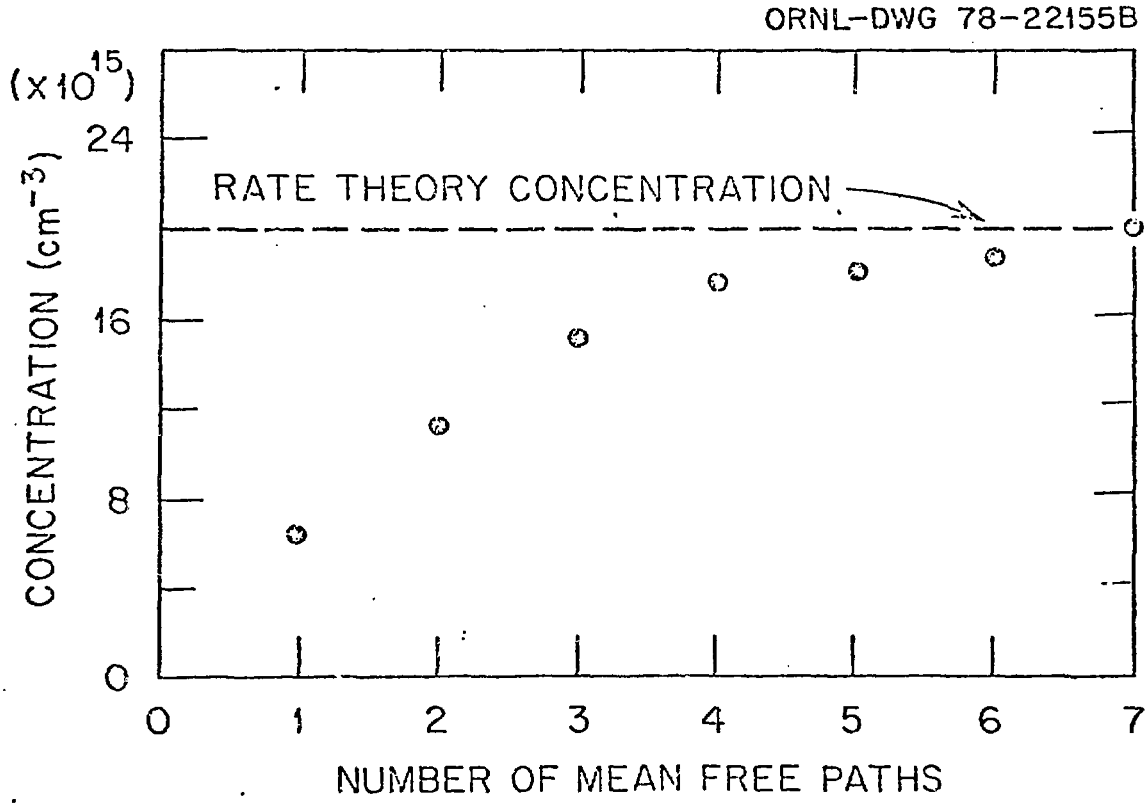
Figure Captions

Fig. 1. Mean free path analysis of the region of material contributing to the concentration at a reference point.

Fig. 2. Vacancy concentration as a function of time at an arbitrary reference point for a steady dose rate of 10^{-6} dpa/s at 500°C and a dislocation density of 10^{11} cm^{-2}.

Fig. 3. Interstitial concentration as a function of time for the same conditions as in Figure 2. Shown is a random time interval spanning three pulses corresponding to three consecutive collision cascades occurring in a steady at various times and distances (numbers at top of figure) from the reference point.

Fig. 4. Fractional difference between the increase in void radius obtained from the cascade-resolved diffusion theory and the rate theory as a function of temperature for the same conditions as in Figure 2.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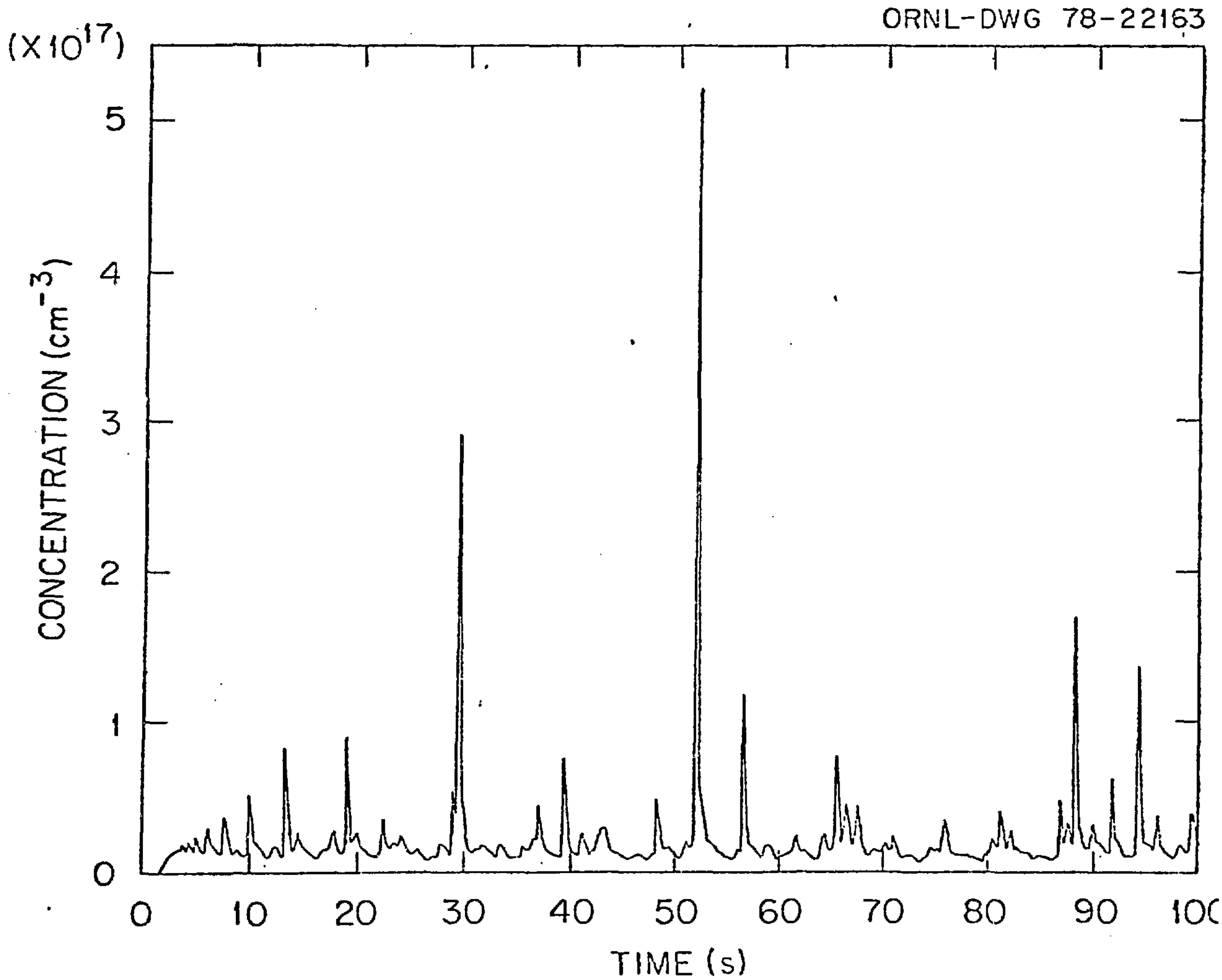
4
5
6
7
NUMBER OF MEAN FREE PATHS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TIME (s)

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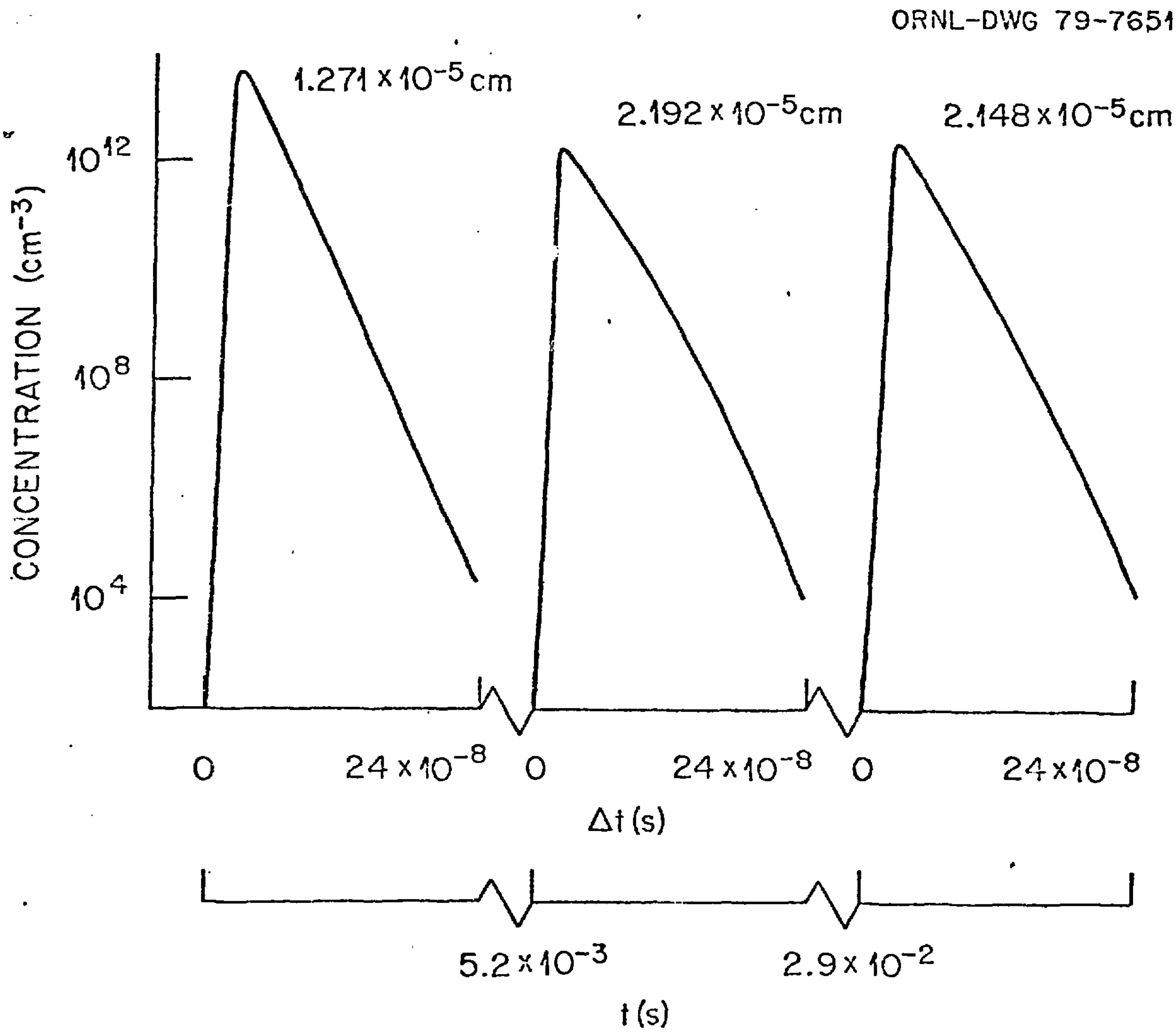
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10^8
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0
24 x 10^-8
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Δt (s)
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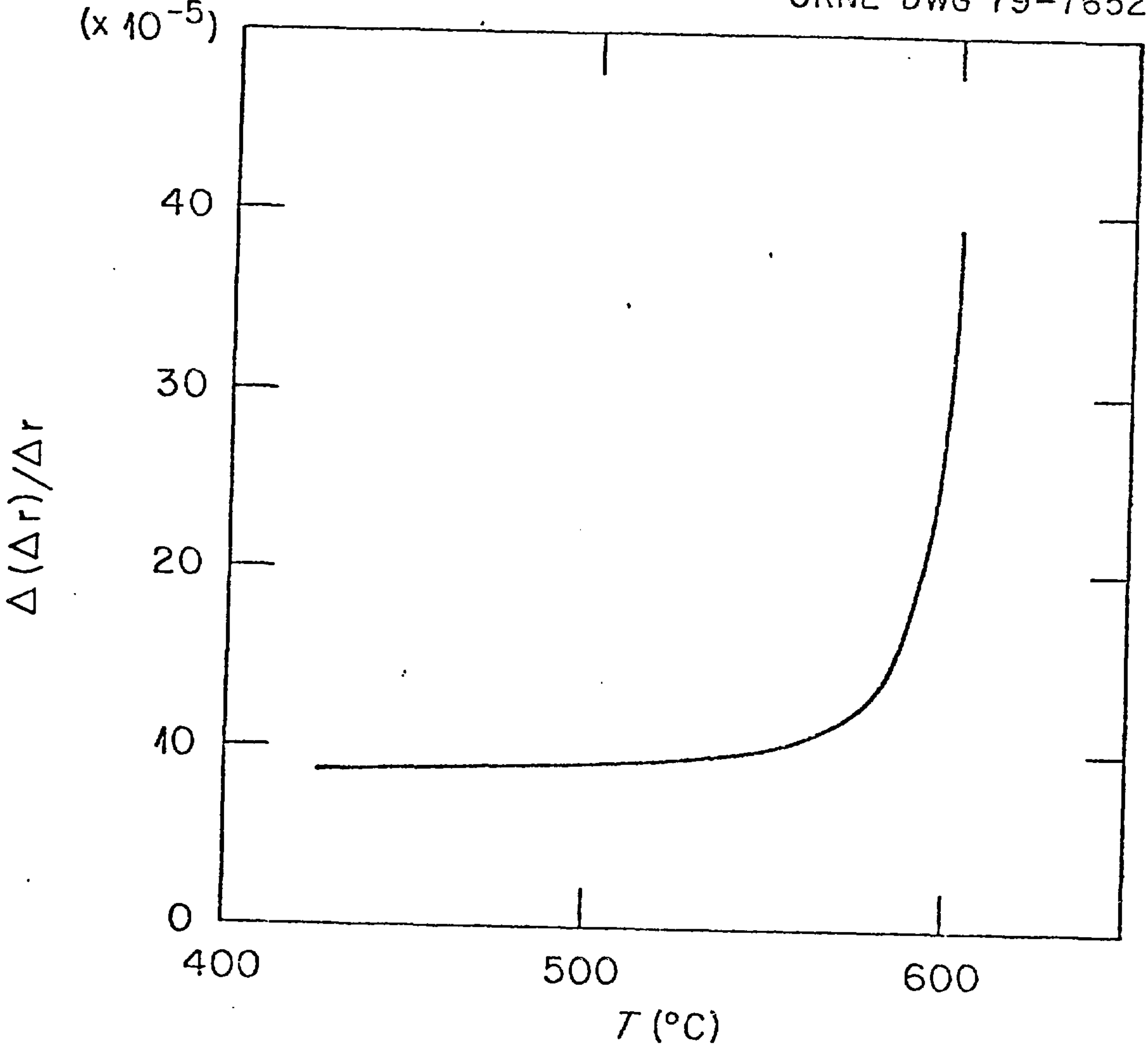
2.9 x 10^-2
t (s)

3

ORNL-DWG 79-7652



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