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Fusion Technology Institute
University of Wisconsin
1500 Engineering Drive
Madison, WI 53706

http://fti.neep.wisc.edu

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N. Ghoniem

G. L. Kulcinski

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Nuclear Engineering Department
University of Wisconsin
Madison, Wisconsin 53706

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Abstract

Theoretical and experimental investigations of materials subjected to a pulsed mode of operation are valuable for a variety of reasons. CTR designs based on a pulsed mode of operation, especially laser fusion reactors, will have to consider such studies. Theoretical and experimental developments in this field will also help to extend understanding of other basic materials phenomena. Experimental evidence has already indicated that materials have a different response to pulsed irradiation as compared to that from steady state irradiations. This report describes a simplified model of void growth under pulsed irradiation. The study shows that the final swelling can be considerably reduced depending on the time shape of the irradiation pulse.

<u>Void Growth Kinetics Under an</u>

<u>Irradiation Pulse</u>

1. <u>Introduction and Scope</u>

Previous studies of void swelling have been focused on the steady state

irradiation conditions. Prime motives for such studies were the development

of breeder cladding materials and materials for fusion power reactors. Materials

and irradiation parameters are known to play an important role in the final

assesment of swelling and mechanical properties. The nature and magnitude of

damage, cascade structure, energy and type of bombarding particle, rate of

damage, gaseous content, impurity content, crystal structure, material composition,

temperature, stress, are all known to affect materials behavior. It is now

also appreciated both on experimental evidence [8-12] and theoretical grounds

[1,2,3,7,15], that the time structure of the irradiation has a considerable

influence on the void growth kinetics.

Theoretical and experimental investigations of materials subjected

to a pulsed mode of operation are valuable for a variety of reasons. The

distinction between steady state analysis and interrupted operation was not

important in the simulation of cladding materials for breeder reactors, but

may play a significant tole in the study of accidents and transient

conditions in fast breeder reactors. On the other hand it becomes crucial in some

CTR first wall materials. Some CTR designs would demand pulsed operation

with the interval between fusion burn cycles ranging from 1 to 10 seconds

(theta pinches and laser reactors) to several hundred seconds (Tokamak reactors)

and pulse widths ranging from nanoseconds (laser reactors) to tens of milliseconds

(theta pinch reactors) to the neighborhood of several thousand seconds (Tokamak

reactors). The fluctuation in temperature, stress and damage rate resulting from pulsed

reactor operation are expected to significantly affect component life. From a fundamental point of view, important parameters in the description of steady state operation can be expected to be measured by pulsed sources such as cyclotrons and intense neutron generators. Hence, theoretical and experimental studies are needed to analyze and understant detailed time-dependent considerations.

2. Problem Status

2.1 Experimental Evidence

Experiments on the production of vacancy condensates by laser bombardment by Smidt and Metz[11,12] showed pecularities special to short-time laser pulses. It was initially believed that the vacancies were formed as a result of quenching, and concentrations as high as 1% (10^2 times that expected at the meltins point) have been observed. The generation of vacancies by non-conservative motion of jogs did not appear feasible because the dislocation density was too low. Interaction of shock waves generated by the laser pulse is suggested as a possible alternative explanation. Taylor, Potter and Wiedersich[8] suggested the classification of pulses into three regimes: (a) pulse duration short compared with interstitial and vacancy lifetimes, (b) pulse duration long compared with interstitial life but short compared with vacancy lifetimes, and (c) pulse duration long compared with the lifetime of both defects. Their experimental conditions corresponded to case (b). Pure Ni was chosen for the target material. Equal beam-on and beam-off times were used in the experiment with a 13-ms beam-on period. Although the void-volume fraction was independent of the time structure of the irradiation, the void microstructures were different. The number density in the pulsed irradiation was between three and ten times smaller than that obtained in the unpulsed experiments. In the pulsed irradiations, voids were heterogeneously distributed

and appeared to have been nucleated along dislocation lines.

At the Naval Research Laboratory (NRL)[9,10], a study was made of the influence of irradiating with tightly-focused rastered ion beams as opposed to using a defocused beam. Again, it was found that the time variance of the irradiation did not alter the magnitude of the void-volume fraction but did affect the size and void density. The effect of rastering on the void-number density and void size were found to be opposite to the previous data obtained with steady state beam.

It, therefore, appears that the supersaturation of vacancies(resulting in the nucleation phenomena) and the void microstructure (growth and annealing kinetics) may depend significantly on the time structure of the irradiation.

2.2 Theoretical Treatments

Choi, Russell and Bement[3] treated the problem of void nucleation applied to transient processes. The procedure was to solve numerically a set of difference equations for the time rate of change of void concentrations. They concluded that even voids above the critical nucleus size may decay after a sharp reduction in damage rate and that the time constant for void decay is much longer than that for void formation. Odette and Myers[15] studied the effect of interrupted irradiations on void nucleation. The kinetic equations of cluster embryo concentration buildup and decay were solved assuming time dependent square wave vacancy and interstitial concentration profiles character-ized by durations of t_p and separated by intervals of t_d. The nucleation behavior in these systems was found to be determined by competition between the buildup of embryos during the burn and the decay of the embryos. In situations where the pulse time t_p is long and the pulse interval t_d short, nucleation rates build up to a large fraction of the normal steady state rates in a few pulses.

In contrast, for short pulses and long intervals nucleation rates only build up to small fractions of steady state rates even after a long number of pulses. Their problem is formulated in terms of equations describing multistate kinetic phenomena Numerical solutions to the set of resulting equations were sought assuming a vacancy concentration $C_v = SC_v^e$ during each pulse, where S is the vacancy super-saturation, and $C_v = 0$ during intervals between pulses. From the study, it appeared that, in some situations of interest to the CTR program, the pulsed nature of the irradiations might result in a negligible rate of void swelling. These situations notably include the low flux, short burn time and small duty cycle conditions anticipated for some fusion test devices and even some conceptual experimental power reactors (EPRs).

Sprague, Russell and Choi[23] investigated the applicability of a steady state and time-dependent homogeneous void nucleation theory to charged particle irradiation experiments The steady-state theory was found to be inappropriate, since significant changes in a metals microstructure would likely occur before the void nucleation rate could reach steady state. Two types of time dependence were examined, that of the point defect concentrations at the beginning of an irradiation and the longer-term time dependence of void nucleation. It was found that vacancy and interstitial clustering significantly reduce the relaxation time for the point defect concentration.

Not only is void nucleation found to be affected by pulsed irradiation, but also void growth and final material swelling is also affected by this method of operation. Schiffgens, Graves and Doran[2] made a study of the effects resulting from damage rate fluctuations, dividing their analysis into three separate parts. The goal of the first part of their analysis is to obtain the probabilities for vacancy and interstitial leakage as functions of dislocation density and temperature. They approximated the random dislocation configuration of a real

solid by an idealized configuration consisting of a bundle of parallel straight dislocations for which end effects are neglected. The coupled, steady state, continuity equations for vacancies and interstitials are solved for the concentration profiles in order to obtain the leakage rates of each zone and relate them to their average concentrations at the dislocation densities and temperatures of interest.

In the second part of the analysis, it is assumed that vacancy and interstitial leakage rates are proportional to the respective average concentrations at all times. This is to be considered as a main assumption that enables separation of spacial and time dependence of defect balance throughout the cycle. The results of the spatially averaged concentrations are solved to obtain the time dependences of these average concentrations.

The third part of the analysis builds on the first and the second. It is assumed that voids growing among the dislocations may be treated simply as growing in the time varying average concentrations resulting from the annihilation leakage competition. The fields and the spatial variations in the vacancy and interstitial concentrations near the dislocations are ignored. The uncoupled continuity equations describing diffusion in the region surrounding the void are solved with the requirement that the average concentrations within the region be equal to the average concentration throughout the material at all times.

In their study they identified four regimes associated with void growth in pulsed systems. They showed that the pulse parameters chosen for reactors and irradiation sources can dramatically alter swelling, and most probably other irradiation effects. For example, if the irradiation pulse is less than about 10 μsec and the interval between pulses is greater than about ten vacancy mean lives, no void growth is possible in stainless steel regardless of temperature, dislocation density and pulse amplitude provided no voids form in displacement cascades which are large enough to survive the time between pulses.

3. Annealing Kinetics

At a given temperature T, the vacancy concentration at the surface of the void, C_v, is given by

$$C_v/C_o = \exp \left\{ \left(\frac{\partial F}{\partial n} \right)/kT \right\} \tag{1}$$

where $\frac{\partial F}{\partial n}$ is the change in energy of the configuration per vacancy emitted, and C_o the equilibrium concentration of vacancies. For a spherical hole in an infinite isotropic solid, that 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 in References (16, 17, and 5); one can write the following expressions:

$$\frac{\partial F}{\partial n} = F_m \Omega \tag{2}$$

where Ω is the atomic volume $\sim b^3$. Here F_m is the mechanical force per unit surface area acting on a vacancy at the void surface.

$$F_m = P + \frac{2\gamma}{r_v} + \frac{\gamma^2}{2\mu r_v^2} - P_g \tag{3}$$

Here P is the hydrostatic pressure, γ, the surface energy, r_v 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:

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

While if Van der Waals law is assumed to hold, one gets (25):

$$P_g = \frac{NkT}{(4/3\pi r_v^3 - aN)} - \frac{bN^2}{16/9\pi^2 r_v^6} \tag{5}$$

where a and b are constants.

Normally, the elastic energy is negligibly small[17], so in the general case we have:

$$F_m = P + \frac{2\gamma}{r_v} - P_g \qquad (6)$$

This formulation is useful in studying the general situation where gas atoms are trapped in voids and where stress waves accompany the damage production. Here it is assumed that no stress waves propagate through the solid and that no gas atoms are contained in the void. Future studies should contain these two features.

4. Theoretical Formulation of the time dependence of materials parameters:

    4.1 Time-dependent Rate Theory Model

The steady-state rate theory model was originally developed by Harkness and Li (1969, 1971)[26] and by Wiedersich (1972)[27] and has been extended and redeveloped by Brailsford and Bullough (1972,1973)[4]. The procedure here is to describe the time-dependent rate theory model in its general case, and then develop an analysis applicable to specific material and irradiation conditions. This is done to preserve the physical picture, as much as possible, away from mathematical complexities.

The idea in a rate theory model is to replace all the discrete sinks in the solid, including the voids, by equivalent continuous distributions of sinks in a continuum. In this continuum, the various sinks are given strengths to insure that the flux of defects to such sinks will be as close as possible to the flux at the actual (geometrical) sinks in the real solid. Replacing the discrete and actual distributions of sinks by an equivalent and idealized continuous distribution will have the obvious advantage of space independence of the problem. The vacancy and interstitial concentrations, and the governing equations for these concentrations now reduce to a simpler pair of time dependent equations.

The simultaneous equations for C_i and C_v, the time dependent interstitial and vacancy concentrations (far from sources and sinks), have the form:

{production rate}-{sink removal rate}-{recombination rate}={concentration rate of change}

(7)

A definition of the following parameters is necessary:

v,i = subscripts referring to either vacancy or interstitial property.

ρ_d = total dislocation density

$\xi_1 = \rho_d^{\ell}$ = irradiation production produced dislocation density in the form of interstitial loops.

$\xi_2 = X_{v,i}$ = bias factors for voids

$\xi_2 = Y_{v,i}$ = bias factors for precipitates

$\xi_2 = Z_{v,i}$ = bias factors for dislocations   (8)

$\xi_3 = C_s$ = void density

$\xi_4 = r_s$ = void radius

$\xi_5 = C_p$ = precipitate concentration

$\xi_6 = r_p$ = precipitate radius

$\xi_{7,8} = K_{v,i}$ = production rates of vacancies and interstitials

$\xi_{9,10} = D_{v,i}$ = diffusion coefficients of vacancies and interstitials

α = recombination coefficient of vacancies and interstitials.

From the definitions given above, an easy analysis would show:

Sticking probabilities per sec of vacancies or interstitials to dislocations =

$$D_{v,i} \, Z_{v,i} \rho_d$$

(9)

Then, (dislocation removal rate) = $D_{v,i} \, Z_{v,i} \, \rho_d \, C_{v,i}$ (10)

Same arguments would hold for voids and precipitates to yield:

(void removal rate)= $4\pi r_s C_s \, X_{i,v} D_{i,v} C_{v,i}$ (11)

and   (precipitate removal rate)= $4\pi r_p C_p \, Y_{i,v} D_{i,v} C_{v,i}$ (12)

It is to be noted that the general bias factors $X_{i,v}$, $Y_{i,v}$, $Z_{i,v}$ will control, to a great extent, the growth kinetics.

The enhanced vacancy generation rate is:

$$K_v = K_i + D_v \{Z_v (\rho_d^{\ell} \bar{C}_{v1} + \rho_d^{n} C_v^{e}) + 4\pi r_s C_s C_v^{e}\} \tag{13}$$

where

$$\bar{C}_{v1} = C_v^{e} \exp [-(\gamma_f + F_{el})b^2/kT] \tag{14}$$

and defines the probability of vacancy emission from a faulted dislocation loop: γ_f is the stacking fault energy and $F_{el}b^2$ is the change in elastic energy of the loop per vacancy emitted. $\bar{C}_{v1}$ is considered small when the stacking fault is present ($\gamma_f \neq 0$) and thus such faulted loops will not easily emit vacancies; however, when the growing faulted loops eventually make contact with each other we expect the loops to unfault and the vacancy emission to suddenly increase ($\bar{C}_{v1} \simeq C_v^{e}$). The last term in equation (13) represents the rate of vacancy emission from the voids and for simplicity the effects of surface tension are neglected.

Now the governing equations for the vacancy and interstitial concentrations far from sinks could be stated in the form:

$$K_i - (D_i C_i Z_i \rho_d + 4\pi D_i C_i X_i r_s C_s + 4\pi D_i C_i Y_i r_p C_p) - \alpha C_i C_v = \frac{\partial C_i}{\partial t} \tag{15}$$

$$K_v - (D_v C_v Z_v \rho_d + 4\pi D_v C_v X_v r_s C_s + 4\pi D_v C_v Y_v r_p C_p) - \alpha C_i C_v = \frac{\partial C_v}{\partial t} \tag{16}$$

These are the most general forms of the equations describing the transient behavior of vacancy and interstitial concentrations.

Theoretical treatment of a general case is rather a formidable task, however, computations could be reduced if a suitable material and irradiation case is chosen. The following treatment will be developed for one specific case, with the hope that it will cast some light on further research features that are of considerable importance. The following assumptions are then made:

(a) High dislocation density of material such that recombination of point defects could be ignored compared to the removal rate by sinks.

(b) The only sinks for point defects are the dislocations initially.

(c) Precipitates concentration is so small that their effect on the absorption of point defects is negligible.

(d) Typically the void density is low such that the mean free path for a point defect absorption by a void is much larger than that for point defect absorption by dislocations.

(e) Producton rate of dislocation loops is small such that the dislocation loop density can be neglected compared to preirradiation dislocation density.

A substantial reduction in mathematical difficulties without loss of generalization would follow from assumption (a). Assumptions (b-e) are justified at the early stages of swelling where material parameters do not change substantially. Assumption (d) would require that:

$$D_{v,i} \; Z_{v,i} \; \rho_d \; C_{v,i} \gg 4\pi r_s C_s X_{v,i} \; D_{v,i} C_{v,i}$$

for its validity. This requirement could be expressed as:

$$\rho_d \gg 4\pi \; r_s \; C_s \qquad . \tag{17}$$

The last condition is fulfilled under most irradiation conditions.

The first assumption, (a) will always be satisfied if $D_{i,v} \; C_{i,v} \; Z_{i,v} \; \rho_d$ $\gg \alpha \; C_i C_v$, or for vacancies only:

$$\rho_d \gg \frac{\alpha C_i}{D_v} \quad , \tag{18}$$

and for interstitials alone:

$$\rho_d \gg \frac{\alpha C_v}{D_i} \tag{19}$$

Equations (17), (18) and (19) reflect the possibility of achieving such a set of circumstances at high dislocation densities. In the same time assumption (e) would simplify the enhanced vacancy generation rate function.

$$K_v \cong K + D_v Z_v \rho_d C_v^e + 4\pi r_s C_s D_v C_v^e$$

to

$$K_v \cong K + D_v C_v^e Z_v \rho_d \tag{20}$$

where K_v is the equilibrium vacancy concentration. Assumptions (a-e) then shift the analysis from general material conditions towards investigating the time demain behavior of main parameters. The present study is one step towards a general model for material behavior in which material parameters that vary with time are described as state variables. The interactions and relative effects between these state variables can be described in phase space as will be shown later.

The two governing equations (15) and (16) will now reduce to the following uncoupled first order linear differential equations:

$$K(t) - D_i Z_i \rho_d C_i(t) = \frac{dC_i(t)}{dt} \tag{21}$$

and

$$K(t) + D_v C_v^e Z_v \rho_d - D_v Z_v \rho_d C_v(t) = \frac{dC_v(t)}{dt} \quad . \tag{22}$$

We will now define the absorption time constants for vacancies and interstitials by:

$$\tau_d^v = 1/D_v Z_v \rho_d \qquad (23)$$

and

$$\tau_d^i = 1/D_i Z_i \rho_d \qquad (24)$$

These time constants give a guide to how long it would take for a point defect to migrate to dislocations from the time of the production of the defect. Equations (21) and (22) now take the form:

$$\dot{C}_i(t) = - \lambda_i C_i(t) + K(t) \qquad (25)$$

$$\dot{C}_v(t) = - \lambda_v C_v(t) + K(t) + \lambda_v C_v^e \qquad (26)$$

where

$$\lambda_{i,v} = 1/\tau_d^{i,v}$$

4.2 State space description of the dynamic system:

The state variables of a dynamic system are the smallest set of variables which determine the state of the dynamic system.[18] If at least n variables $\xi_1(t), \xi_2(t), \ldots, \xi_n(t)$ are needed to completely describe the behavior of a dynamic system, then such n variables $\xi_1(t), \xi_2(t), \ldots, \xi_n(t)$ are a set of state variables. These n vairables could be described to be the components of a state vector $\bar{\xi}(t)$. In equation (8) a set of state variables $\xi_i(t)$, (i = 1,2,....,10) are defined. As one would notice here the set of state variables $\xi_i(t)$ are interdependent and, in principle, their change with time could be described.

Mathematical drudgery could be greatly relieved if one were successful to linearize this dynamic system. Consider the multiple-imput-mulitple-output system defined in equation (8). In this system, $\xi_i(t)$ represent the state variables; $U_i(t)$ denote the input variables which could be variables like

the temperature, pressure and concentration of Frenkel pairs. The system
equations would be as follows:

$$\dot{\xi}_1 = a_{11}(t)\, \xi_1 + a_{12}^{(t)}\, \xi_2 + \ldots + a_{1n}(t)\, \xi_n + b_{11}(t)\, U_1 + b_{12}^{(t)} U_2 + \ldots + b_{1r}(t) U_r$$

$$\dot{\xi}_2 = a_{21}(t)\, \xi_1 + a_{22}(t)\, \xi_2 + \ldots + a_{2n}(t)\, \xi_n + b_{21}(t)\, b_{22}(t) U_2 + \ldots + b_{2r}(t) U_r \qquad (28)$$

$$\dot{\xi}_n = a_{n1}(t)\xi_1 + a_{n2}(t)\xi_2 + \ldots + a_{nn}(t)\, \xi_n + b_{n1}(t)\, U_1 + b_{n2}(t) U_2 + \ldots + b_{nr}(t) U_r$$

In terms of vector-matrix notation, these n equations can be written
compactly as

$$\dot{\underline{\xi}} = \underline{A}(t)\, \underline{\xi} + \underline{B}(t)\, \underline{U} \qquad (29)$$

where $\underline{\xi}$ is the state vector and $\underline{U}$ is the input vector. The solution of
equation (29) is given in equation (30)[19]

$$\bar{\xi}(t) = e^{\int \underline{A} dt}\, \bar{\xi}(\theta) + \int e^{\int \underline{A} dt}\, \underline{B}(t')\, \underline{U}\, dt' \qquad (30)$$

4.3 Analysis of a delta function generation of point defects

In actual circumstances the production function of point defects is
dependent on the source of irradiations, the duty factor of the bombarding
particle. The real assesment of the spatial and time dependence of the
production rate function $K(t)$ is the subject of a future study related to
fusion reactors. As a point of this study, a delta function generation of
point defects will be assumed.

$$K(t) = \varepsilon K_o\, \delta\, (t-t_p) \qquad (31)$$

where εK_o is the strength of the δ-function and t_p is the time at which
generation takes place, ε being the small time interval during which a constant
generation of point defects at a rate of K_o dpa/sec takes place.

The rate of change of the void radius is given as:

$$\frac{dr_v}{dt} = \left(D_v \frac{dC_v(t)}{dr} - D_i \frac{dC_i(t)}{dr} \right)_{r=r_v} \tag{32}$$

Certain assumptions regarding the previous equation were extended to the present analysis:

(i) The diffusion coefficients near the void surface is the same as for the bulk.

(ii). Concentration gradients at the void surface are constants within a capture distance from the surface of the void and change to zero at larger distances. A capture distance of 4 lattice parameters is used based on Beeler's Simulation Studies[20], Wolfer continuum approach[21] and Bullough's Work[22].

Equation (32) can now be written as:

$$\frac{dr_v}{dt} = \frac{1}{\ell} (D_v C_v - D_i C_i) - \frac{D_v C_v^e}{\ell} \exp (F_m b^3 / kT) \tag{33}$$

where F_m is the mechanical force acting on the void surface defined by equation (3), and ℓ is the capture distance.

In this analysis no attempt was made to include the effects of external stress or gas pressure. Also since the changes in the void radius would be small for one pulse, the mechanical force was taken as a constant depending on the initial void size as:

$$F_m = \frac{2\gamma}{r_v^o} \tag{34}$$

It is noticed that equations (25), (26) and (33) are in the state variable form with the vector $\underline{\xi}(t)$ having its components $C_v(t)$, $C_i(t)$ and $r_v(t)$ only.

Combining the 3 equations together will give:

$$\frac{d}{dt}\begin{bmatrix} C_v \\ C_i \\ r_v \end{bmatrix} = \begin{bmatrix} -\lambda_v & 0 & 0 \\ 0 & -\lambda_i & 0 \\ \frac{D_v}{\ell} & -\frac{D_i}{\ell} & 0 \end{bmatrix}\begin{bmatrix} C_v \\ C_i \\ r_v \end{bmatrix} + \begin{bmatrix} K(t)+\lambda^v C_v^e \\ K(t) \\ -\frac{D_v}{\ell}C_v^e e^{(F_m b^3/kT)} \end{bmatrix} \tag{35}$$

This system is easily solved with the aid of equation (30) and for a delta driving function to give the following expressions:

$$C_i(t) = K_o \varepsilon\, e^{-\lambda(t-t_p)} U(t-t_p) \tag{36}$$

$$C_v(t) = C_v^e + K_o \varepsilon\, e^{-\lambda(t-t_p)} U(t-t_p) \tag{37}$$

$$r_v(t) = r_v^o + \frac{K_o \varepsilon}{\ell} U(t-t_p)\left\{ \frac{D_v}{\lambda_v}(1 - e^{-\lambda_v(t-t_p)}) - \frac{D_i}{\lambda_i}(1 - e^{-\lambda_i(t-t_p)})\right\}$$

$$- \frac{D_v C_v^e}{\ell}(\exp(F_m b^3/kT)-1)t \tag{38}$$

where $U(t-t_p)$ is the unit step function at t_p.

It is appropriate at this point to define two times relevent to the kinematic behavior of a pulse, the recovery time (t_r) and the pulse annealing time (t_a) as follows:

$t_r \equiv$ The time at which the void regains its initial radius.

$t_a \equiv$ The time at which the void loses the increace in its volume due to the pulse, i.e., when the void returns back to its original radius. These two markers are shown in Fig. 1.

With reasonable approximations[*] it is possible to derive the following

simple expressions for t_r and t_a:

$$t_r = \tau_v \ln \frac{Z_v Z_i}{(Z_v - Z_i)} \tag{39}$$

$$t_a = K\varepsilon (\frac{1}{Z_v} - \frac{1}{Z_i})/\rho_d D_v C_v^e (\exp(F_m b^3/kT)-1) \tag{40}$$

The final swelling of the material might be expected to be dependent

on the time between pulses and the annealing kinetics under current conditions.

An analytical expression of the conditions required for the recovery time

to equal to the pulse annealing time could then be helpful in determining

the response of blanket materials to pulsed irradiation. If the pulse starts

at time $t = 0$, then such conditions are expressed as:

$$\frac{K\varepsilon e^{E_v^f/kT}}{(e^{F_m b^3/kT}-1)} = \frac{Z_i}{(Z_i - Z_v)} \ln \frac{Z_i Z_v}{(Z_i - Z_v)} \tag{41}$$

4.4 Steady State Irradiation Versus Pulsed Irradiation

It is interesting to compare the case of steady state irradiation with

a similar pulsed irradiation condition.

If one supposes that a constant rate of point defect production is

present in the material, then the previous formulation would give the variation

of void radius with time as:

$$r_v(t) = r_v^o + \frac{(Kt)}{\ell \rho_d} (\frac{1}{Z_v} - \frac{1}{Z_i}) \tag{42}$$

---

[*] It is readily seen that at times near t_r, interstitial and annealing effects
can be neglected. While for times in the range of t_a, no time dependent
vacancy and intersitial contributions have to be considered.

This result shows a linear dependence of the void radius on the accumulated dpa as was obtained by Bullough and Perrin for high dislocation densities[4].

For a number of pulses accumulating the same dpa as the previous case of constant irradiation , for temperatures low enough that no annealing commences and for periods between pulses greater than t_r, one would get the same expression (42) and would have the same amount of swelling as a comparable case of constant irradiation.

5. Results and Discussions

5.1 General Time-Behavior of Voids

Fig. (1) shows the general behavior of a void radius as time proceeds, based on the previous model. It is seen from this figure that the dynamical response of the void radius to the pulse exhibits a variety of features.

The time domain behavior of the void radius is devided into five regimes as shown in Fig. (1) for a void with an initial radius of 500 Å, dislocation density $5 \times 10^{11}/cm^2$ and at a temperature of 700°C.

Parameters used in this calculation for Ni and are given in Table (1).[4]

(i) Interstitial Migration Stages

Immediately after the irradiation pulse, the interstitials start migrating to the sinks (dislocations in this case) and to the void. For the early stage of irradiation, the flux of interstitials to the void is higher than the flux of vacancies which then results in a decrease in void size. The high velocities of interstitials responsible for this effect can be easily seen from Fig.(2) where it is evident that they essentially disappear after a few intersitial life times (about 10^{-8} seconds in this case).

(ii) First equilibrium stage

After most of the interstitials disappear from the system, there is a time period ($\sim 10^{-7}$ sec in this case) during which little happens to the void. The vacancies

have not arrived at the void surface in significant numbers to cause a noticeable size increase.

(iii) Vacancy Migration Stage

Following the first equilibrium stage, vacancies begin to migrate into the void with appreciable quantities giving rise to an increase in the void size. Once the irradiation produced vacancy concentration drops to very low values, no further changes in the void radius occurs. This is completed in about a micro-second for this case.

(iv) Second Equilibrium Stage

In this stage the irradiation produced vacancies and interstitials cease to exist in appreciable amounts and because the interstitials have been perf-erentially absorbed by the dislocations, the void size increases. It is to be remarked that if the metal is irradiated during this stage, a net increase in swelling will be the result. If the system is pulsed during any of the four other stages, no increase in swelling over that in the first pulse would be expected. Further work on conditions satisfying that is under development.

(v) Annealing Stage

At elevated temperatures vacancies are effectively emitted from the void surface. As described in section (3) the emission probability of a vacancy from the void surface is primarily affected by the mechanical force in the solid. If we assume that no forces other than the surface tension are acting on the vacancy at the void surface, then it is apparent that the void will shrink. It is also noted that it takes a rather long time to anneal out the original void, especially at low temperatures. Subsequent work is intended to address the question of void stabilization by the presence of gas atoms and its effect on pulsed systems.

5.2 Temperature Effects

An increase in the temperature of the system will increase the mobility of both vacancies and interstitials, and dynamical process will take place at an earlier time. This is depicted in Fig. (4) where annealing kinetics begin to play an important role at higher temperatures.

It is noticed that voids anneal much quicker for higher temperatures due to the increase in the vacancy emission probability from the void surface with temperatures.

Figures (2) and (3) demonstrate the effect of temperature on the average vacancy and interstitial concentrations. The combination of a high equilibrium vacancy concentration value, at elevated temperatures, and a small void radius predict that such void will anneal even more rapidly than a large void as shown in Figure (5). However, it must be re-emphasized that we have not accounted for any gas atoms in the void which might reduce the shrinkage rate of the void.

5.3 Consequences of Changes in Dislocation Density

Dislocations have been assumed to be the dominant sink. As the dislocation density goes up the residual point defects have less effect on the void kinetics as shown in Fig. (6). In the second equilibrium stage, which corresponds to steady state swelling, the higher dislocation density actually lowers the increase in void size. This is also realized in the steady state operation, where it was postulated, and later confirmed, that swelling could be minimised by cold working the material.[24]

Lower dislocation densities would affect the transient and second equilibrium stages as seen from Fig. (6). This conclusion stems from the time behavior of point defects as dislocation density changes. It is

seen from Figures (7) and (8) that at any time the average concentration of interstitials and vacancies is higher with a lower dislocation density. This will lead to an enhanced effect of point defects on transient stages. It is interesting also to note that final higher swelling is attained in the second equilibrium stage for lower dislocation densities, in accordance with experimental results[24].

5.4 Recovery and Annealing Times

Recovery and annealing times were defined in section (4-3) and it is relevant to examine how effective material and irradiation conditions can alter those two times. Since the final swelling is determined by the time interval between pulses, the width of the stable window from the recovery time to the annealing time could be important in pulsed systems. The difference between the annealing time and the recovery time, called the stable window width, is plotted in Figure (9) versus temperatures for different dpa values injected into the pulse.

It is seen that the stable window width decreases as the temperature goes up and as the dpa rate goes down.

In Figure (10) the stable window width is seen to be affected by the dislocation density. It goes up with dislocation density especially at lower temperatures while at higher temperatures the effect is negligible.

6. Conclusions and Expected Achievements

The model and the assumptions considered in this work are simple. It is not intended for this work to analyze general conditions of materials and pulse shape and reach performance conclusions of pulsed systems. The main objective rather is to clarify possible differences from steady state operation

and to identify probable time domain regimes. Also sensitivity to major materials conditions and irradiation parameters were kept in mind throughout the work.

The likeliness of having different kinetics characteristics in different time regimes might have a large impact on irradiation conditions to get favorable materials performance.

Even if nucleation of voids were found to be the same, pulsing the system in any regime other then the second equilibrium regime could result in a final decrease in swelling. So it is important to link growth and nucleation models under such pulsed conditions.

Future research will assume more realistic directions in the light of this sensitivity study. It is important to include other materials parameters (i.e. precipitates, irradiation produced loops, presence of gas atoms, etc.) in this study. Recombination studies together with a better treatment of gradient problems near the void surface would certainly lead to a broader perspective. Competition of other sinks with dislocations as a means of point defect removal will be valuable in the assessment of highly damaged material. It is realized also that realistic conditions will have to include pressure waves and temperature transients accompying the pulse. Mainly annealing is non-linear, and a train of pulses with a period between pulses greater than t_r will have to include this non-linearity to get a reasonable value of final characteristics after a reasonable number of pulses.

In conclusion, the growth model has to be more sophisticated and to be tied up with other models (in particular nucleation) to allow a final judgement that swelling can be dramatically reduced by suitably chosen materials and irradiation parameters.
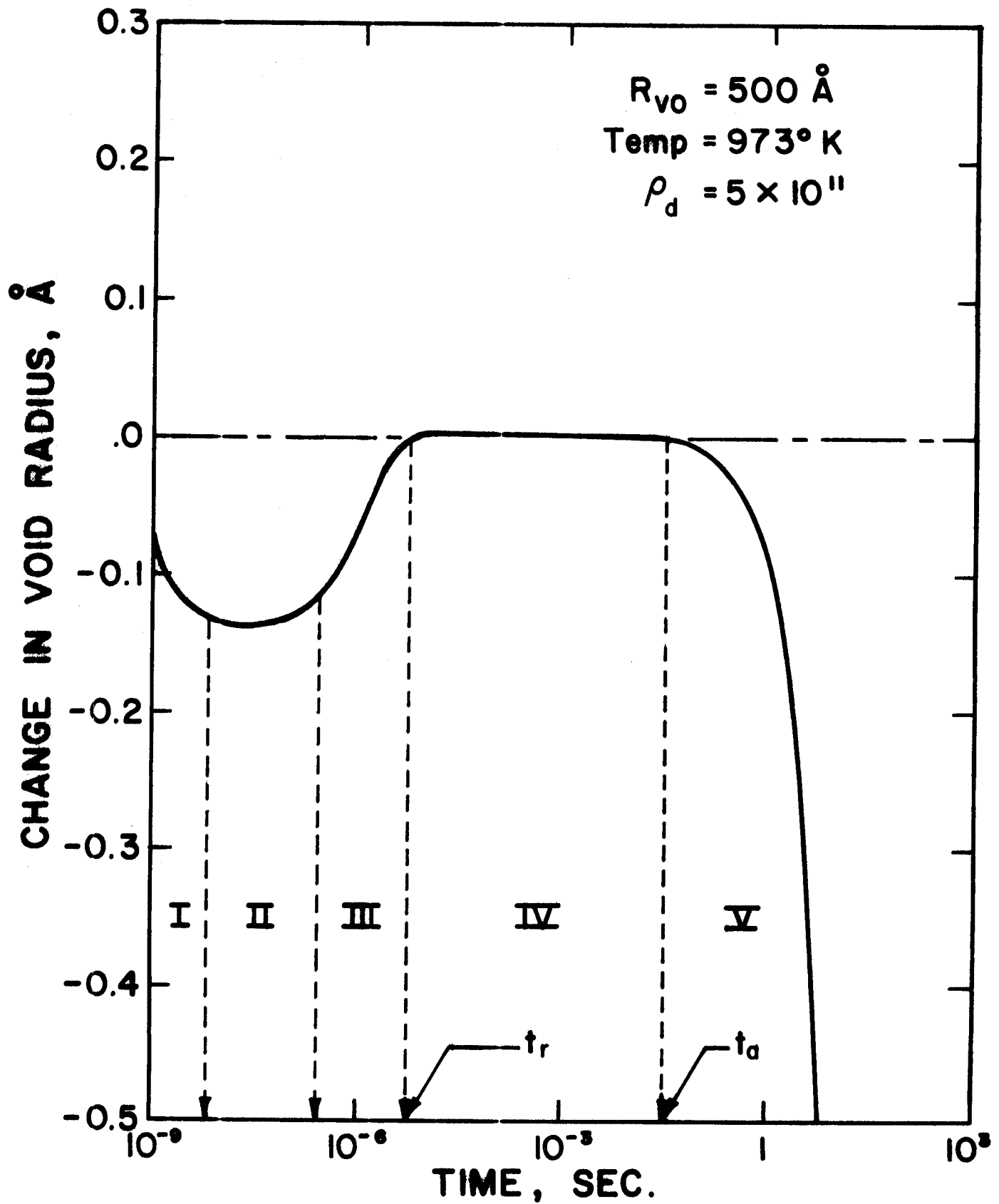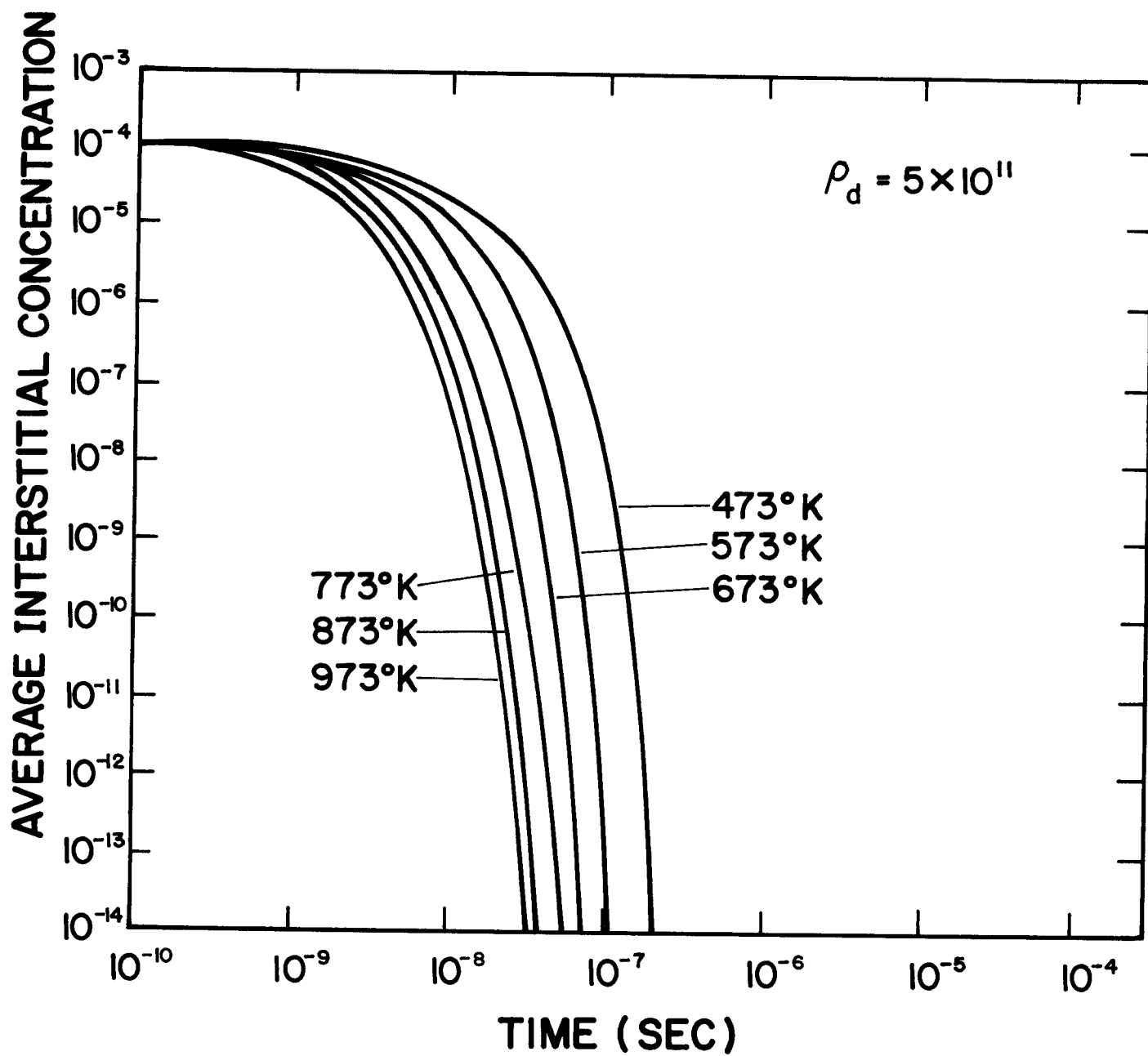
Fig. I   Change in void radius after a pulse.

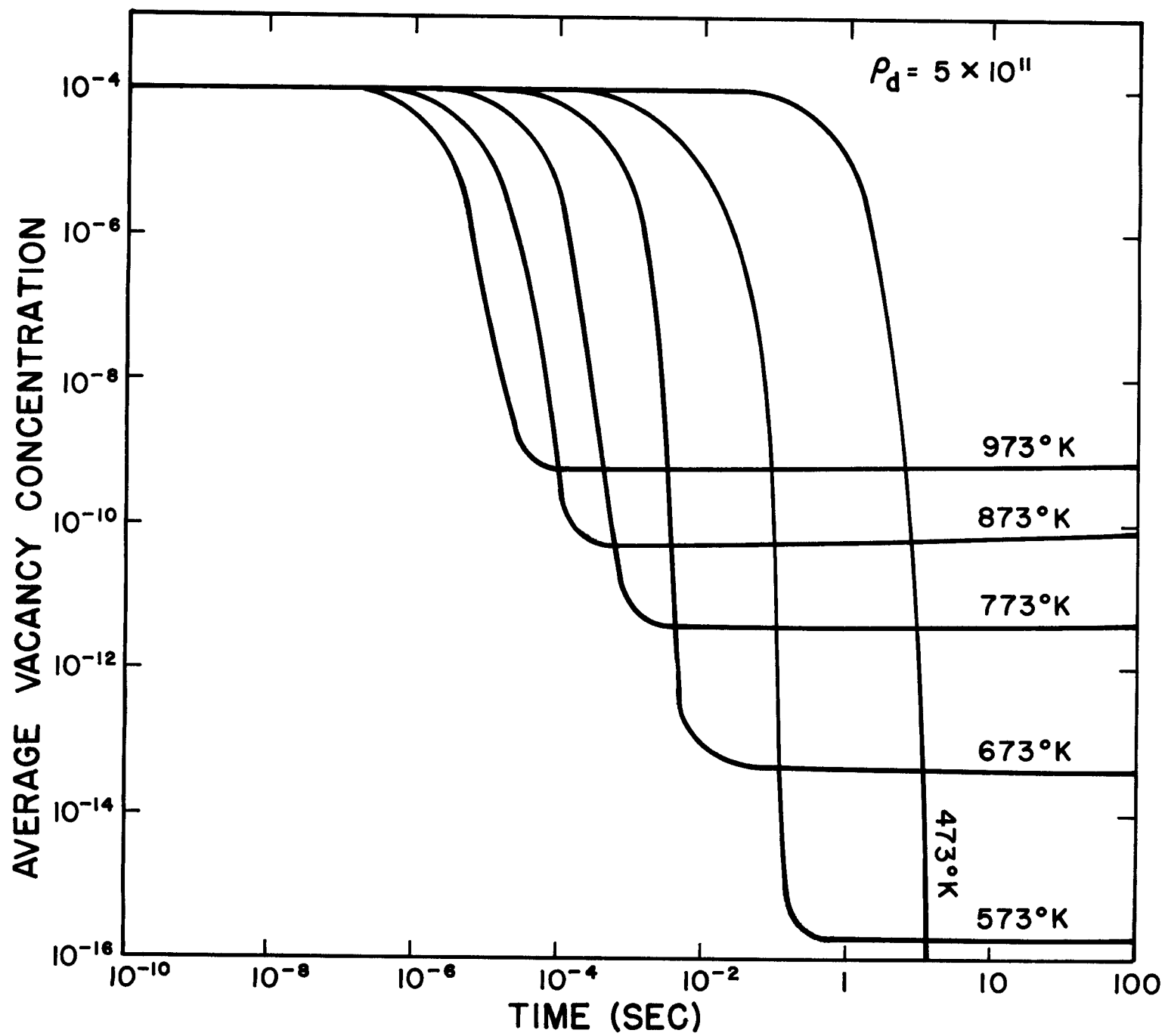Fig. 2 Effect of temp. on average interstitial concentration.
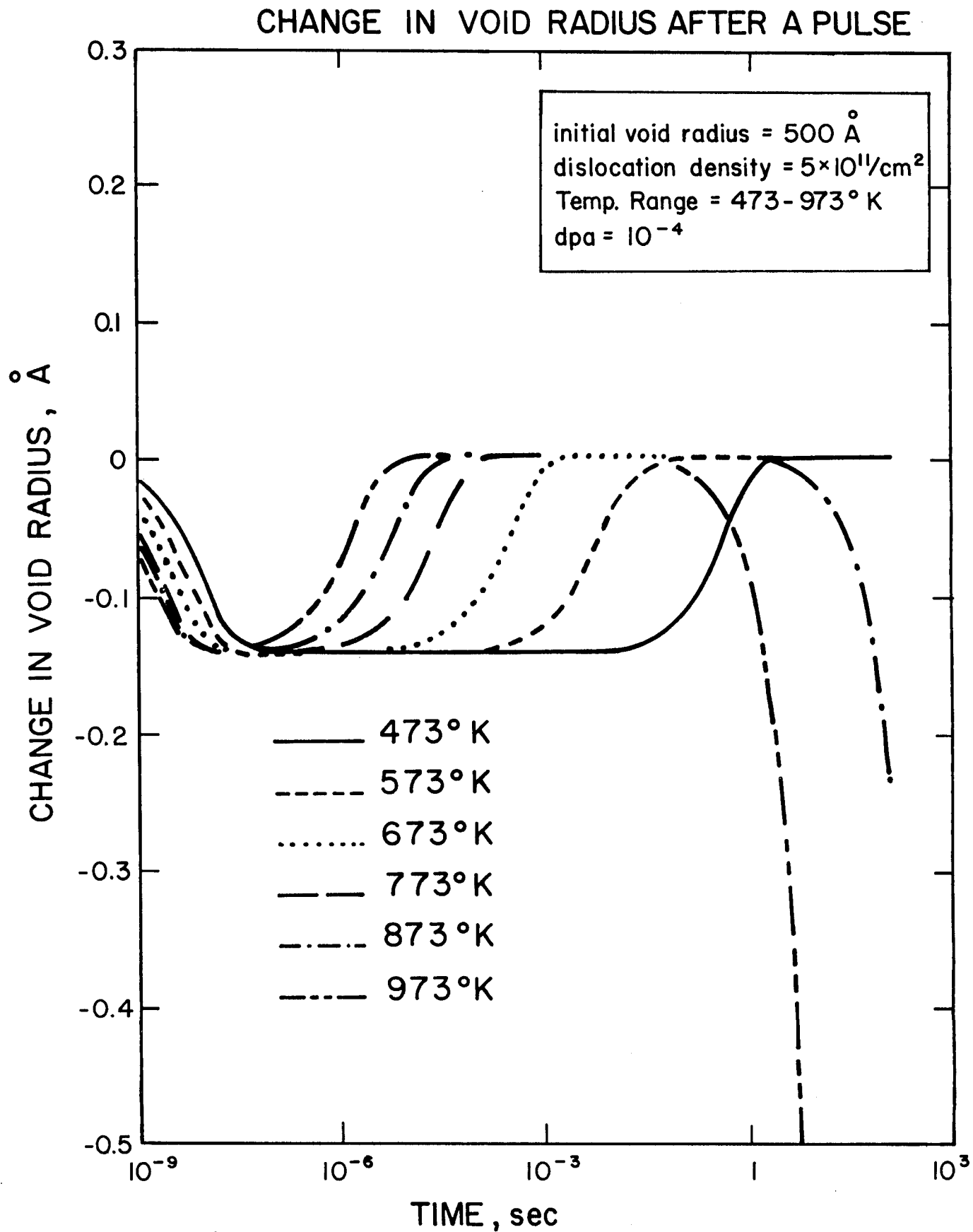
Fig.(3)   Effect of temp. on average vacancy
concentration

CHANGE IN VOID RADIUS AFTER A PULSE

FIG. 4   EFFECT OF TEMPERATURE ON VOID KINETICS FOR A LARGE VOID
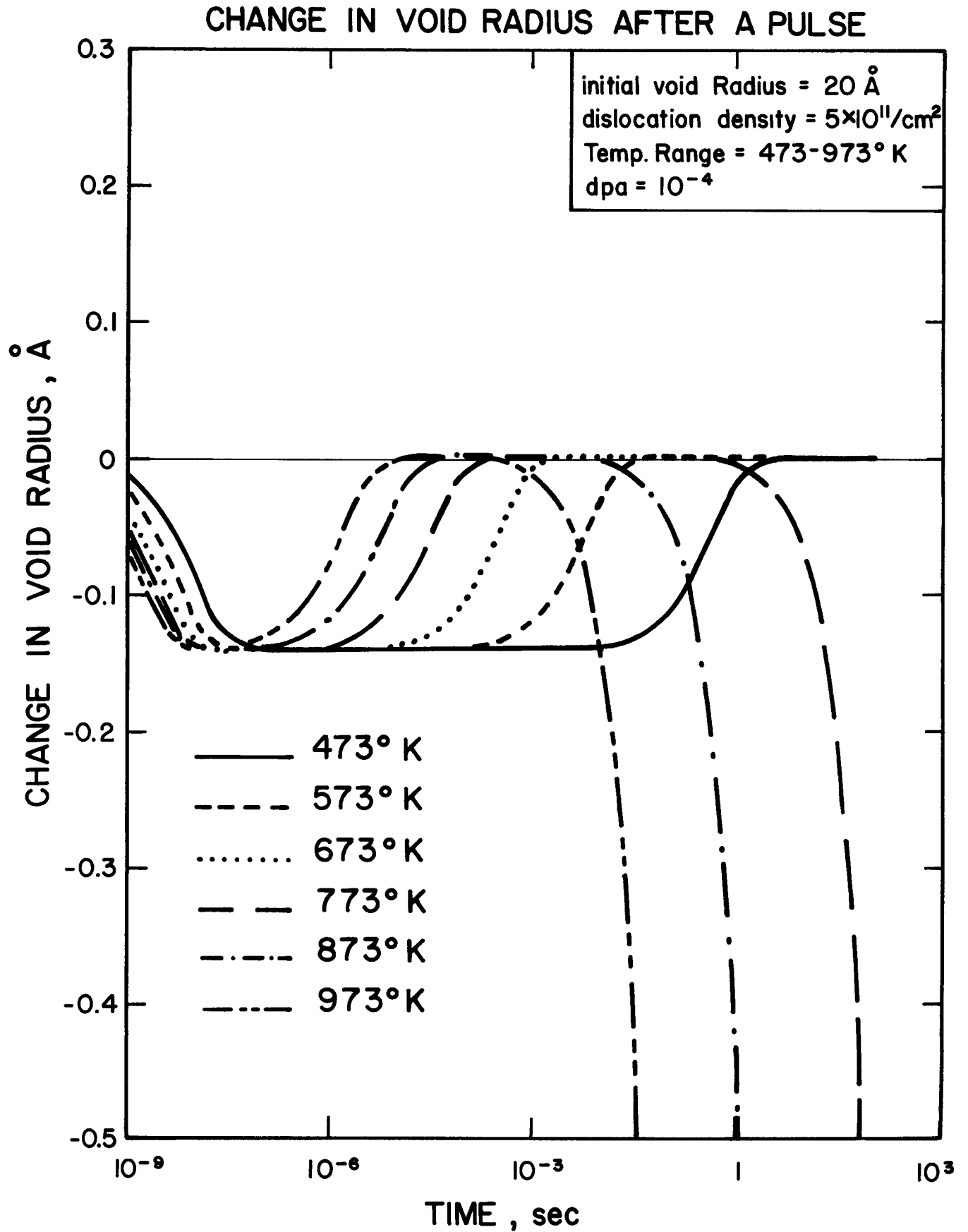
CHANGE IN VOID RADIUS AFTER A PULSE

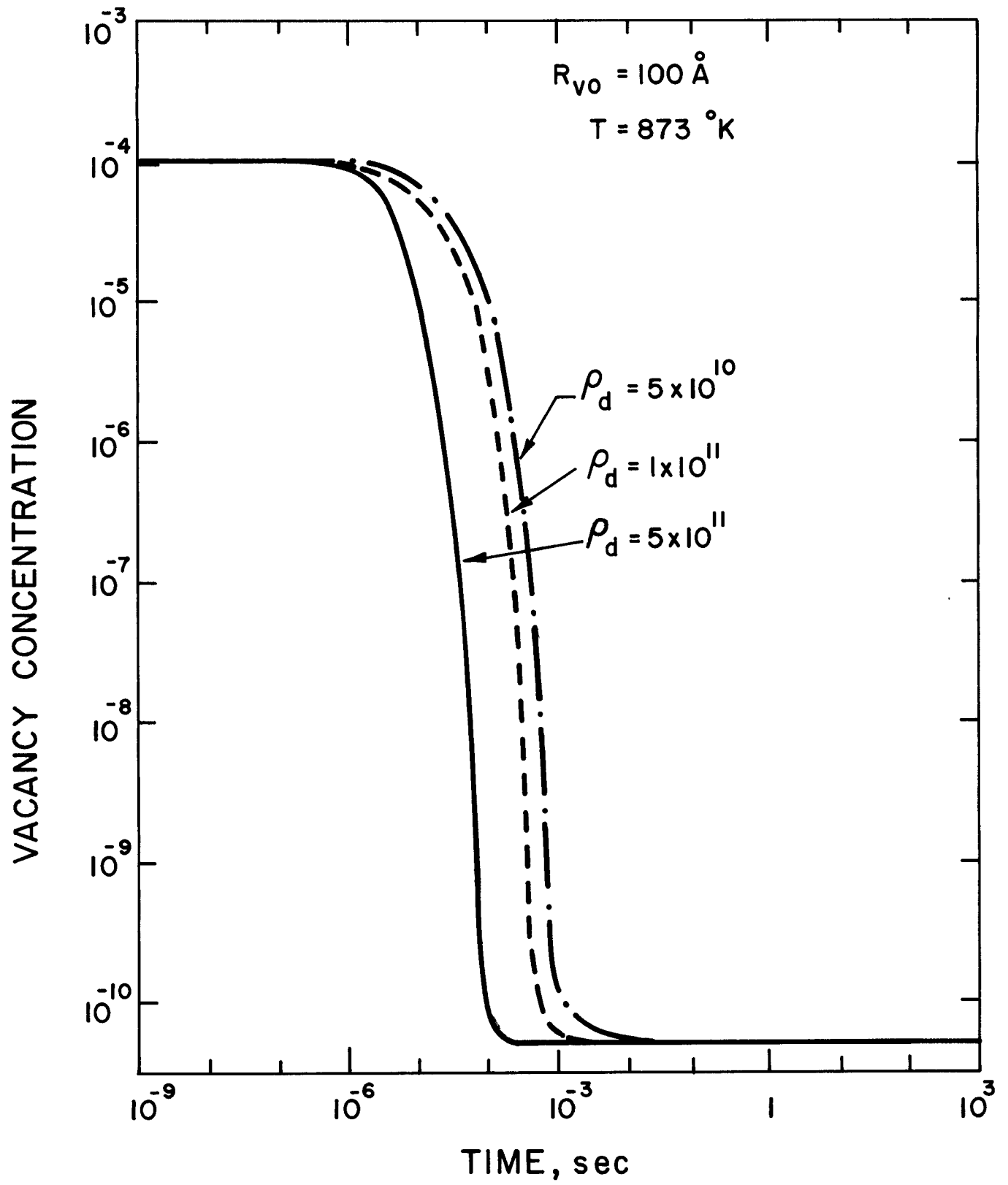FIG. 5 Effect of temperature on void kinetics for a small void
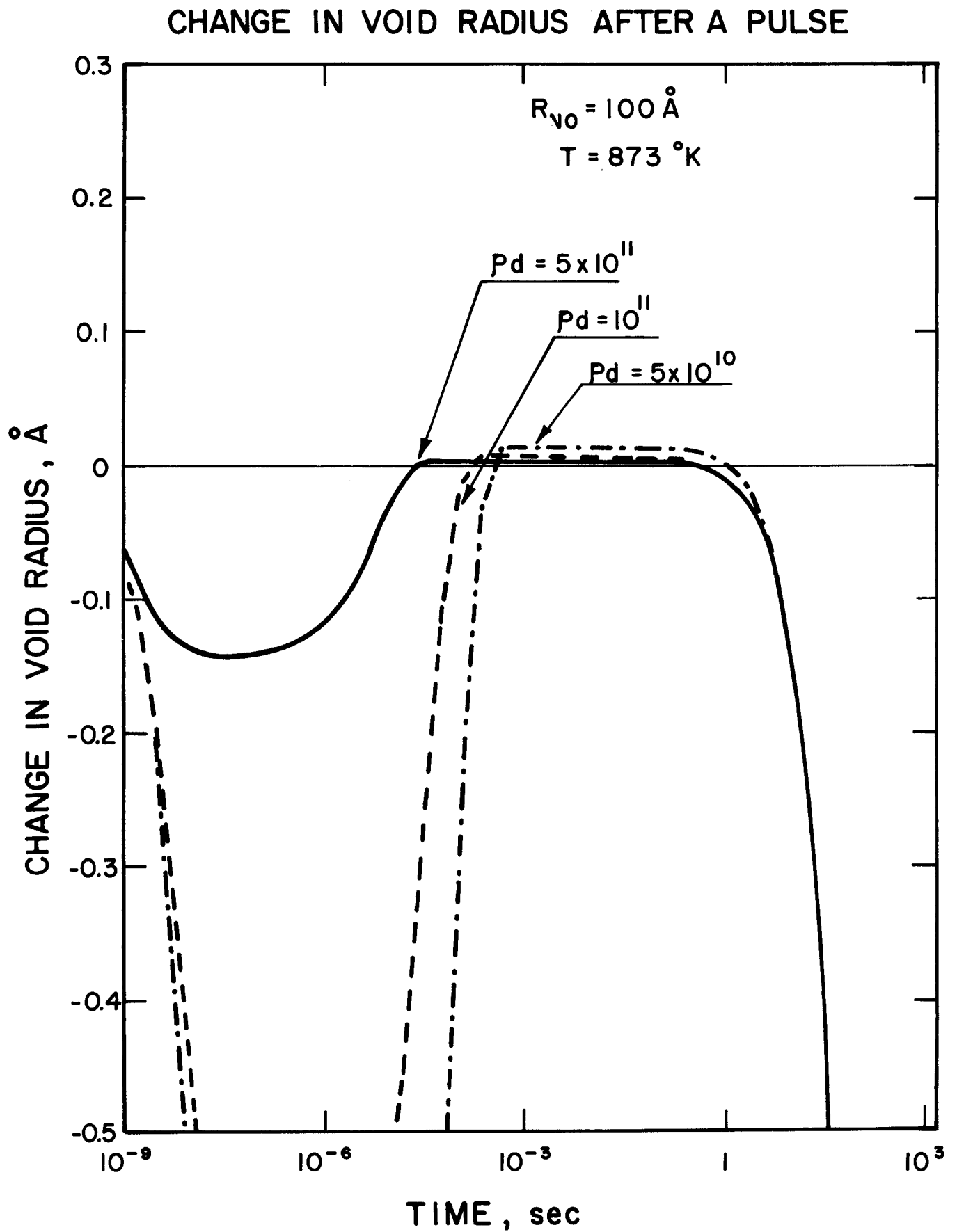
FIG. 6. EFFECT OF DISLOCATION DENSITY ON VOID KINETICS

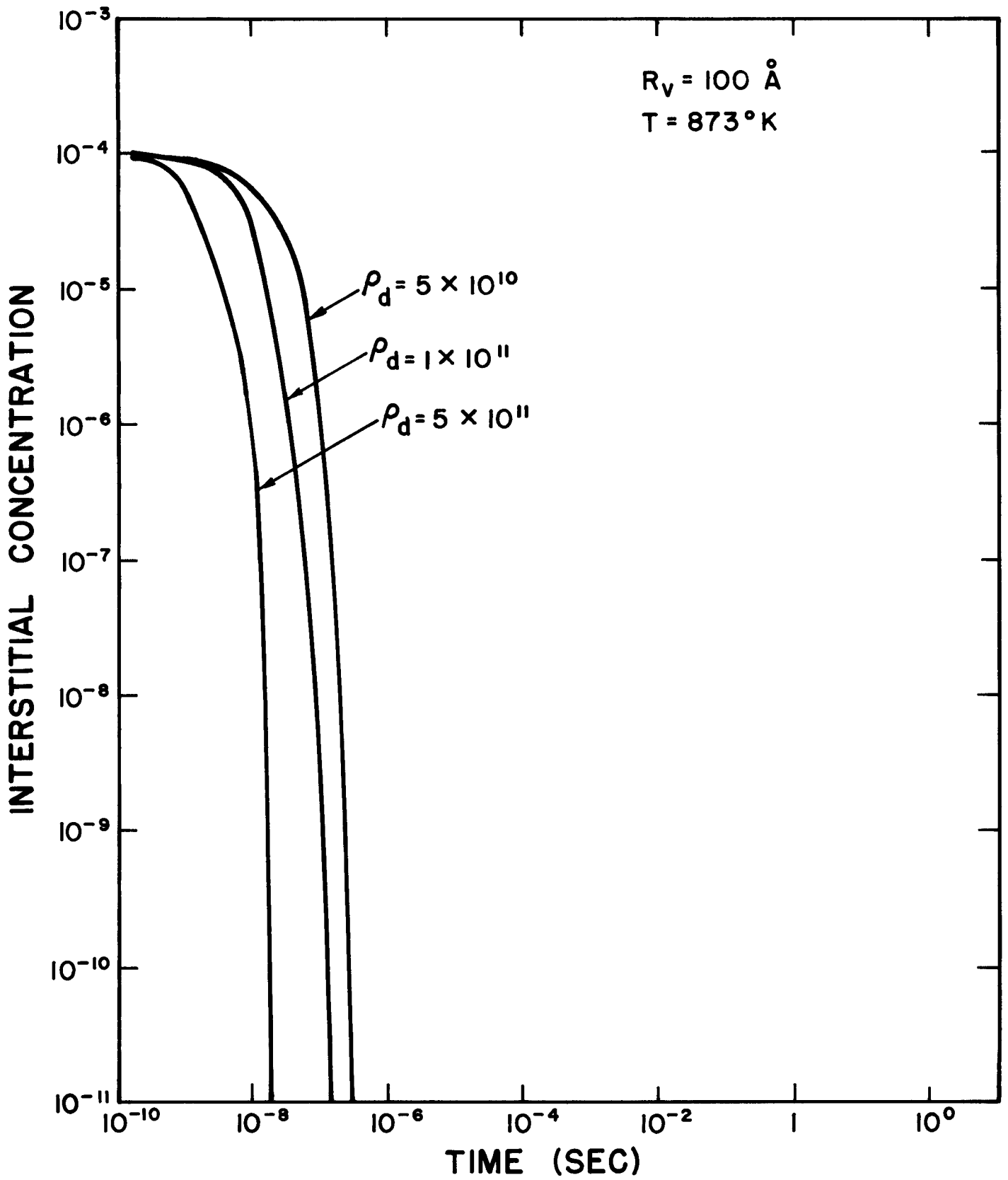FIG. 7. EFFECT OF DISLOCATION DENSITY ON C_v

Fig.(8)   Effect of dislocation density on Ci
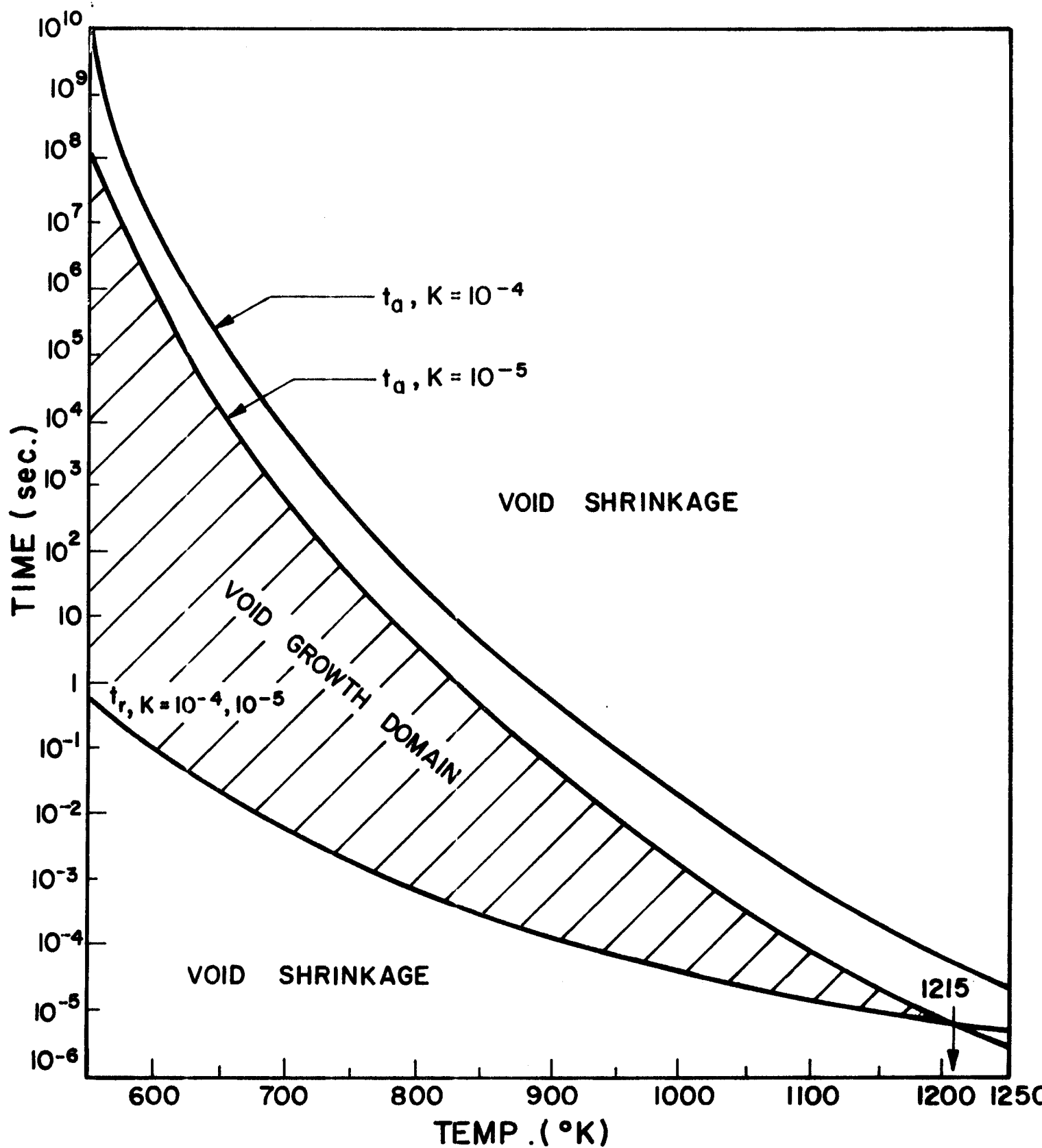
FIG.(9) EFFECT OF TEMPERATURE ON RECOVERY AND
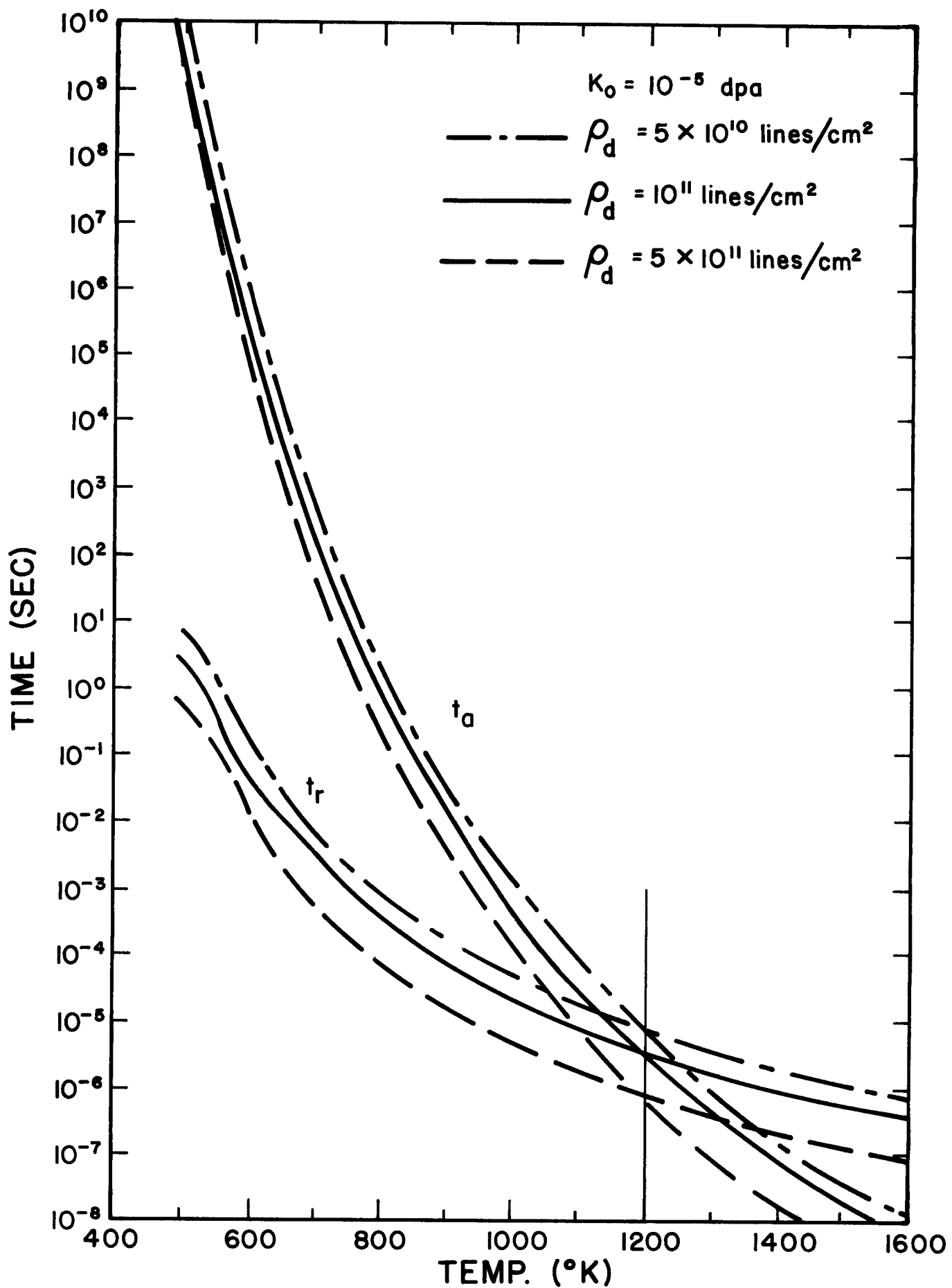ANNEALING TIMES.

Fig. (10)   Effect of dislocation density on equilibrium period

TABLE I (4)

Assumed Parameters for Nickel

E_v^f	:	Vacancy formation energy (eV)	1.77
E_i^f	:	Interstitial formation energy (eV)	4.00
D_v^o	:	Vacancy pre-exponential facter (cm^2/sec)	0.19
D_i^o	:	Interstitial pre-exponential factor (cm^2/sec)	0.008
E_v^m	:	Vacancy migration energy (eV)	1.00
E_i^m	:	Interstitial migration energy (eV)	0.15
b	:	Lattice parameter (Å)	3.5
γ	:	Surface free energy (eV/cm^2)	6.2415 x 10^{14}
Z_v	:	Bias factor for vacancy	1.00
Z_i	:	Bias factor for interstitial	1.01
K	:	Total damage in the pulse (dpa)	10^{-4}

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