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CASCADE-INDUCED FLUCTUATIONS AND THE TRANSITION FROM THE STABLE TO THE CRITICAL CAVITY RADIUS FOR SWELLING*

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ABSTRACT

Recently, a cascade diffusion theory was developed to understand cascade-induced fluctuations in point defect flux during irradiation. Application of the theory revealed that such fluctuations give rise to a mechanism of cascade-induced creep that is predicted to be of significant magnitude. Here we extend the investigation to the formation of cavities. Specifically, we explore the possible importance of cascade-induced cavity growth excursions in triggering a transition from the gas-content-dictated stable radius to the critical radius for bias-driven growth. Two methods of analysis are employed. The first uses the variance of fluctuations to assess the average effect of fluctuations. The second is based on the fact that in a large ensemble of cavities, a small fraction will experience larger than average excursions. This prospect is assessed by estimating upper limits to the processes. For the conditions considered, it is concluded that cascade-induced fluctuations are of minor importance in triggering the onset of swelling in a population of stable gas-containing cavities.

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1. INTRODUCTION

In a material subject to energetic neutron or heavy-ion irradiation, knock-on events lead to point defect generation in cascades. Previous theoretical work by one of us and coworkers [1,2] has demonstrated the enormous magnitudes of the resulting spatial and temporal fluctuations in local point defect concentrations and fluxes. Since the formation and behavior of extended sinks for point defects depends on these fluxes, it is reasonable to ask whether the overall microstructure, and the properties to which it gives rise, are influenced by fluctuations.

Subsequently, a new mechanism of irradiation creep, cascade-induced creep, was revealed by this work and shown to be of large magnitude [3,4]. On the other hand such fluctuations were shown to have relatively little effect on the growth of large cavities, when compared to the swelling results predicted by a conventional rate theory picture where fluctuations are not considered [1]. In related work it was also shown that an averaged rate theory approach is valid for the growth of cavities in a periodically varying environment that does not include cascade-induced fluctuations, i.e., where the irradiation is pulsed in time but spatially uniform [5,6].

In the present paper we address the possible importance of cascade-induced fluctuations on the early stages of cavity formation. This work is based on the existence of a critical radius above which cavity growth is driven by the dislocation-cavity bias. Where gas is present in a material, the existence of both stable radii (smaller than the critical radius) and critical radii have been established theoretically and experimentally [7–11]. The theory leading to these results is a quasi-steady state rate theory where point defect generation is modeled as continuous in time and space; fluctuations are neglected by definition. However, it is obvious in principle that fluctuations in the point defect flux at a cavity may induce growth or shrinkage excursions. The question explored here is whether such excursions could be of sufficient magnitude to be important in transferring cavities from the stable to the critical cavity radius. We thus pre-suppose an existing population of gas-containing cavities.

2. ACHIEVING BIAS-DRIVEN GROWTH

2.1 Stable and Critical Cavity Radii

In continuum rate theory the growth rate of a cavity in a material undergoing irradiation may be expressed as [9]

$$\frac{dr_c}{dt} = \frac{\Omega}{r_c}\,[Z_v^c D_v C_v - Z_i^c D_i C_i - Z_v^c D_v C_v^e] \;, \qquad (1)$$

where r_c and t denote cavity radius and time. The quantity Ω is the atomic volume, the $Z^c_{v,i}$ are capture efficiencies of the cavity for vacancies and interstitials, and C_v^e is the thermal equilibrium vacancy concentration at a cavity of radius r_c. The quantities $D_{v,i}$ [$= D^o_{v,i}\exp(-E^m_{v,i}/kT)$] are the point defect diffusion coefficients, where $D^o_{v,i}$ are pre-exponential constants, $E^m_{v,i}$ are the corresponding point defect migration energies, and kT has its usual meaning. The

$C_{v,i}$ are the bulk-averaged concentrations of point defects, expressions for which are given in numerous references in terms of materials parameters and irradiation conditions (see, for example [9]). The values of the quantities used in subsequent calculations are given in Table 1.

The strengths of sinks for point defects dictate their concentrations. Here the only sinks considered are dislocations and cavities. The total sink strength is thus $S_{v,i} = S^c_{v,i} + S^d_{v,i}$, where the superscripts c and d denote cavities and dislocations. These are expressed as

$$S^c_{v,i} = 4\pi r_c N_c Z^c_{v,i} \ , \qquad (2)$$

and

$$S^d_{v,i} = L \ Z^d_{v,i} \ , \qquad (3)$$

where N_c is the cavity concentration, L is the dislocation density and $Z^d_{v,i}$ are the dislocation capture efficiencies for piont defects. Higher order sink corrections [12] are ignored and, for the numerical evaluations, all Z's are set to unity except Z^d_i, which is the parameter used to embody the dislocation-cavity bias. The concentration C^e_v is given by,

$$C^e_v = C^o_v \exp[-(P_g - 2\gamma/r_c)\Omega/kT] \ . \qquad (4)$$

Here

$$C^o_v = \Omega^{-1} \exp(S^f_v/k)\exp(-E^f_v/kT) \qquad (5)$$

is the bulk thermal equilibrium value, and S_v^f and E_v^f are the entropy and enthalpy of vacancy formation. The pressure of contained gas in the cavity is P_g and the surface free energy is γ. For order-of-magnitude estimates in the range of interest here, it is sufficient to relate P_g to the number of contained gas atoms, n_g, through the ideal gas law

$$P_g = \frac{3\ n_g\ kT}{4\pi r_c^{\ 3}} \ . \tag{6}$$

The effects of more complex equations of state in the evaluation of critical quantities are treated elsewhere [8,9].

The interpretation of Eq. (1) is quite simple in principle. For a given set of conditions and contained number of gas atoms, the balance between the net radiation-induced influx of vacancies and the thermally emitted outflux of vacancies determines whether or not a cavity of a given radius will grow. It has been shown that a full interpretation provides a convincing picture of many aspects of cavity growth [9]. Some examples are the existence of a temperature shift of swelling with dose rate, a dose interval to the onset of swelling, and the appearance of bimodal cavity size distributions.

Representative solutions are shown in Fig. 1. For n_g less than a critical number n_g^*, there are two physically meaningful solutions for the condition $dr_c/dt = 0$. The lower root, r_c^s, is denoted as the stable radius. It is that radius where, without fluctuations, a cavity would reside in the steady state. It is larger than the thermal equilibrium radius corresponding to n_g, because of the excess radiation induced vacancy flux [9]. The larger root, r_c^c, is designated as the critical radius. It represents the size above which the cavity undergoes continued growth during irradiation. Also shown are the maximum critical radius, r_c^o, corresponding to $n_g = 0$, and the minimum critical radius r_c^*, corresponding to $n_g = n_g^*$. Where a cavity contains more than n_g^* gas atoms, no critical radius exists; that cavity grows inexorably by bias-driven growth. Figure 2 shows that above a certain temperature range, r_c^* (and correspondingly, n_g^*) increases very rapidly with temperature.

Thus a stable cavity can enter the regime of rapid growth by two qualitatively different processes. The first is by continued accumulation of gas, produced by transmutation or injection, for example, to achieve n_g^*. Without fluctuations the time to the onset of rapid swelling is the time to the accumulation of n_g^*. The alternative path depends upon fluctuations to bring cavities normally residing at r_c^s, and containing $n_g < n_g^*$ gas atoms past the size r_c, whereupon they continue to grow. In any system there are natural fluctuations from various processes. Here we investigate the possibility of cascade-induced cavity growth excursions.

2.2 Bridging the Gap Between r_c^s and r_c^c

As shown in Fig. 1, r_c^s approaches r_c^c as n_g approaches n_g^*, and the gap that fluctuations must bridge decreases. We first establish the required number of vacancies corresponding to the difference between r_c^s and r_c^c as a function of n_g and T. This is accomplished by solving Eq. (1). Under the conditions specified it is found that $n_g^* = 345$ and $r_c^* = 1.42$ nm. The results for r_c^s and r_c^c vs n_g are shown in Fig. 3, for n_g near n_g^*. Shown also are the number of vacancies, Δn, needed for conversion corresponding to several values of n_g. For example, with $\Delta n_g \equiv n_g^* - n_g$ we obtain: $\Delta n_g = 1$, $\Delta n = 69$; $\Delta n_g = 2$, $\Delta n = 212$; $\Delta n_g = 15$, $\Delta n = 775$.

The cavity is only likely to be converted when the magnitude of cascade-induced fluctuations is significant with respect to Δn. Figure 4 shows the dependence of Δn versus temperature on n_g. Also shown is the variation of n_g^* with temperature. This curve bounds the variation of Δn with temperature, since for $n_g > n_g^*$ neither r_c^s, r_c^c, nor the gap between them exists. Thus Fig. 4 also reflects the dependence of Δn upon Δn_g. It is evident that the vacancy gap per gas atom rises exponentially with temperature. For example, calculations at 690°C give $n_g^* \sim 10^5$ and $\Delta n \sim 10^6$ for $\Delta n_g = 1$. Thus we expect that fluctuations are much less potent at triggering conversions at high temperatures.

3. ACHIEVING CRITICAL RADIUS BY CASCADE-INDUCED FLUCTUATIONS

3.1 Cascade Diffusion Theory

In cascade diffusion theory the point defects generated in each cascade are tracked by diffusion equations with discrete source terms.

The point defect flux falling on a sink such as a cavity is the superposition of contributions arising from cascades that have occurred at all earlier times in the entire material volume. Thus it is found that even during a steady irradiation there are extreme fluctuations in the defect flux from one instant to another at a given point and from one point to another at a given time. The instantaneous current from a single cascade to a cavity of radius r_c is [2]

$$I_c = \frac{\upsilon\, r_c(R-r_c)}{2(\pi D\tau^3)^{1/2}R} \quad \exp[-(R-r_c)^2/4D\tau]\exp(-DS\tau) \ . \tag{7}$$

Here υ is the number of point defects of either kind in the cascade, R is the distance of the cascade center from the cavity center, D is the point defect diffusion coefficient and $\tau = t - t_c$, where t is the time of observation and t_c is the (earlier) time at which the cascade occurred. S is the sink strength given by the sum of Eqs.(2) and (3). Superposition of solutions of this type for a typical neutron irradiation of nickel gives the profile of vacancy concentration shown in Fig. 5. Large fluctuations are evident.

In our subsequent analysis, several additional relationships derived from Eq. (6) are useful. We denote by n_c the number of defects collected over all time by a cavity from a single cascade. This is given by

$$n_c = \frac{r_c}{R} \exp[-S^{1/2}(R - r_c)] \ . \tag{8}$$

We also need the total number of defects collected by a sink up to time t from all cascades, as well as the variance in this quantity. The necessary expressions are derived in Ref. [2]. For present purposes the ratio of the variance to the mean is sufficient. Denoting the former as σ^2 and the latter as $\bar{n}_c$, we may write

$$\frac{\sigma^2}{\bar{n}_c} \approx \frac{S^{1/2}}{2(1 + S^{1/2} r_c)} \qquad DSt >> 1 . \tag{9}$$

The above results are used in the two subsequent sections to assess the importance of fluctuations. Two kinds of estimates will be used to make the evaluation.

3.2 Average Fluctuations

When a cavity has grown to size r_c^s, we may identify this with $\bar{n}_c$, the average number of vacancies collected up to time t,

$$\bar{n}_c = \frac{4\pi}{3\Omega} r_c^{s\,3} . \tag{10}$$

The additional number of vacancies required to reach r_c^c is thus

$$\Delta n = \frac{4\pi}{3\Omega} r_c^{c\,3} - \bar{n}_c . \tag{11}$$

From Eqs. (9), (10), and (11) we obtain,

$$\frac{\sigma}{\Delta n} = \left[\frac{S^{1/2} r_c}{2\bar{n}_c(1+S^{1/2} r_c^s)}\right]^{1/2} \left[\left(\frac{r_c^c}{r_c^s}\right)^3 - 1\right]^{-1} \tag{12}$$

This last equation compares the average size of fluctuations in point defect accumulation to the size of the gap necessary to convert stable cavities to growing cavities. The relationship is plotted in

Figs. 6 and 7. In Fig. 6 the ratio is shown as a function of cavity radius r_c^s for a range of dislocation densities often encountered in irradiation experiments. All curves are for a difference, $r_c^c - r_c^s$, of 0.1 nm. We note that the rms fluctuation is equal to or less than about 3% of the mean number collected. For high dislocation densities a maximum occurs at about 1 nm. The relative size of fluctuations is thus very small even for radial excursions as small as 0.1 nm. Figure 7 shows the ratio as a function of $r_c^c - r_c^s$ for the same dislocation density range and a cavity radius, r^s, of 2 nm. Again, the ratio is very small except for $r_c^c - r_c^s \ll 0.1$ nm.

3.3 Upper Limits

As in the previous section, we consider the effects of fluctuations in vacancy flux only. Ignoring interstitial accumulation tends to maximize the size of the effect. On average, of course, vacancy accumulation at cavities is only slightly larger than interstitial accumulation as dictated by continuing gas accumulation and by the bias. If, even in this approximation, the effect of fluctuations is small then in reality it must be smaller still.

The analysis can be simplified further by making other assumptions that maximize the effect. Equation (8) when multiplied by υ, the number of vacancies in a cascade, gives the number of vacancies absorbed by a cavity from a cascade over all time. For simplicity, assume both that all these defects are delivered instantly and that $\upsilon = 1000$, a high value corresponding to a cascade initiated by a fusion reactor neutron or to a high energy transfer from an incident ion.* If $n_c > \Delta n$ then a single cascade is capable of triggering a conversion. However, vacancies from a sequence of n cascades, each delivering n_c^i defects, may be necessary,

$$\sum_i^n n_c^i \geq \Delta n \ . \qquad (13)$$

These would occur over a time interval t. The physical significance of t is discussed below. Integration of Eq. (8), weighted by R^2, over all space shows that neither nearby nor distant cascades contribute many defects on average--the nearby ones because they are so few and the distant ones because the point defects are absorbed in the intervening lossy medium. The region of space contributing most lies at $R = S^{-1/2}$. Simplifying again by assuming that all cascades contribute as if at $R = S^{-1/2}$, and keeping in mind that fluctuations can only be significant for small cavities ($S^{1/2}r_c \ll 1$), we rewrite Eq. (8) as

$$n_c = \frac{\upsilon S^{1/2} r_c}{e} \qquad (14)$$

The probability of n or more cascades in volume V in time interval t is [2]

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*A cascade of such large size would probably form in several subcascades. However, they would be correlated in time and for a distant sink would appear in their delivery of point defects much like a single cascade.

$$p_n(t) = \sum_{m=n}^{\infty} P(m,\lambda t) , \tag{15}$$

based on the Poisson probability for exactly m events

$$P(m,\lambda t) = \frac{(\lambda t)^m}{m!} \exp(-\lambda t) . \tag{16}$$

Here λ is the mean rate of cascade occurrence in the volume V, given by

$$\lambda = GV/\upsilon , \tag{17}$$

where G is the point defect production rate in physical units. Since it has been shown that more than 90% of the defects arise from cascades occurring within a distance of $5S^{-1/2}$ of the sink [2], we again simplify by taking V as the volume of that radius. Eqs. (14) - (16) can be used to evaluate the probability that a specified number of vacancies could be supplied within a time interval t.

Figure 8 shows the Δn required as a function of Δn_g. The probability of achieving Δn in a time interval t can be obtained from $p_n(t)$ the probability of n or more cascades, by translating the number of cascades n to the number of defects collected through Eq. (13) and (14). This is shown as a function of Δn in Figs. 9 and 10. Figure 9 is for short time intervals, $t \leq 1$ s, and $\Delta n_g \leq 20$, while Fig. 10 goes up to 21 and 1400, respectively.

Relevant time intervals are appreciated as follows. If the probability were 10^{-6}/s, then to be certain of a transfer of that stable cavity one would have to wait for 1 dpa at a typical reactor dose rate of 10^{-6} dpa/s. However, in this same time interval a cavity in a typical microstructure would accumulate ~100 additional helium atoms at a generation rate of 5 appm/dpa. Thus, referring to Figs. 8 and 10, we see that the additional vacancy accumulation necessitated in response to the gas atom accumulation would be larger than the vacancy contribution in a maximized fluctuation caused by a sequence of cascades.

Figure 11 shows, in an alternative way to Fig. 4, the number of gas atoms that can be deficient, Δn_g, together with n_g^* as a function of temperature. If the Δn_g is above the line shown, then it is not possible to produce a conversion even by a sequence of cascades. It is seen, for example, that at 900 K the cavity must have accumulated all but ~0.2% of n_g^*, the number of gas atoms required for spontaneous conversion, in order to be within range for conversion by cascade-induced fluctuations.

To connect these estimates to a more easily measurable quantity during an experiment, we have made approximate calculations of the time to the onset of swelling with and without fluctuations. As an upper limit estimate, we assume that fluctuations can trigger swelling as soon as enough gas is accumulated to bring Δn_g to the values shown in Fig. 11. Here for simplicity we assume that all the gas is trapped at cavities. The cavity density is taken to be temperature dependent. The values used are those reported by Farrell [13]. Figure 12 shows both the relative and absolute values of the reduction in dose. Above 893 K the two doses are essentially identical. However, below 1 dpa any difference in the dose required would probably be unimportant. Thus any effect could only be discernible over a very small temperature range and even then it is small.

4. DISCUSSION

Cavity growth above the critical size is observed experimentally. In view of the above results indicating small effects of fluctuations, we conclude that the achievement of critical cavity size is driven by gas accumulation.

Cascades may also influence the formation of cavities by the enhanced production of three-dimensional clusters. This is certainly a process by which, in the presence of gas, the initial cavity embryos could be formed. It has been shown previously (see for example [14]) that the nucleation rate depends on the square of the vacancy concentration. During fluctuations C_v^2 may increase enormously above the average (Fig. 5). With contained gas, the clusters will not be destroyed by the corresponding interstitial fluctuations. There will also be enhancement because of the direct generation of three-dimensional vacancy clusters in cascades. Inclusion of such a mechanism in a rate theory picture using discrete clustering equations [15] is possible and would allow us to explore the significance of such events. Thus, we expect that the existence of cascades also affects the number densities of cavities.

Based on previous work where we found that cascades lead to an important new mechanism of irradiation creep [3], and on the present work where the effect on cavity transitions was found to be minimal, criteria can be discerned regarding where cascades may be important in ion beam or neutron irradiation processes. These are that the triggering event must depend on point defect fluxes from not more than a few cascades and that the triggered process must be irreversible so that fluxes of the opposite point defect type do not cause a negating back reaction.

Mechanisms involving phase changes during irradiation, for example, should be examined in this connection.

5. SUMMARY

The influence of cascade-induced fluctuations in point defect flux on swelling, through an effect on cavity formation, has been investigated. Concepts of stable and critical cavity radii established in earlier theoretical and experimental work are applied. The stable radius is that at which a cavity containing a given number of gas atoms resides. It does not grow on the average. For each contained number of gas atoms below a critical number, there also exists a critical radius larger than the stable radius. When a cavity either achieves the critical radius or absorbs the critical number of gas atoms, it will grow inexorably. As the number of gas atoms approaches the critical number from smaller values, the gap between the stable and critical radii narrows.

In the investigation reported here we examined the possibility that a cavity may make excursions from the stable to the critical radius through cascade-induced fluctuations in point defect flux. Two methods were used. Both are based on cascade diffusion theory, an approach developed recently wherein the discrete production of point defects in cascades is accounted for. The first method is based on a derived expression for variance in the number of point defects collected at a cavity. This reflects an average behavior of the cavity population. It is found that the variance is generally small with respect to the number of vacancies needed to produce an experimentally significant excursion from stable to critical radius. The second method is a very approximate evaluation of upper limits and corresponds to the largest excursions that a very few cavities in the population may experience. Here again it is found that cascade-induced growth excursions produce differences that are experimentally insignificant with respect to the case where fluctuations are ignored. It is therefore concluded that the main mechanism underlying the achievement of bias driven cavity growth is gas accumulation.

Criteria are suggested by which it may be judged whether cascade-induced fluctuations may be important in ion beam or neutron irradiations.

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Table 1. Parameter values used in the calculations

$\Omega(m^3)$	$D_v^o(m^2/s)$	$E_v^m(eV)$	$D_i^o(m^3/s)$	$E_i^m(eV)$	$r_r(m)$	$\gamma(J/m^2)$	$L(m/m^3)$	Z_i^d	$E_v^f(eV)$	$G(dpa/s)$
1.095×10^{-29}	1×10^{-6}	1.4	1×10^{-5}	0.15	4×10^{-10}	1	5.10^{14}	1.05	1.6	10^{-6}

FIGURE CAPTIONS

1. Schematic of cavity growth rate as a function of cavity radius for typical reactor dose rates and temperatures. Quantities describing cavity radius and contained number of gas atoms are defined in the text.
2. Behavior of the critical radius as a function of temperature. Solid curve shows the gas-free critical radius and dashed curve shows the minimum critical radius where the cavity contains the (temperature dependent) critical number of gas atoms.
3. Variation of the stable and critical radii as a function of the contained number of gas atoms showing the range near n_g^* and r_c^*.
4. Variation of Δn with temperature for several values of n_g. Also shown is the variation of n_g^* with temperature. This curve bounds the solutions for Δn, since for $n_g > n_g^*$ no critical radius exists.
5. Vacancy concentration as a function of time as calculated by cascade diffusion theory.
6. The ratio $\sigma/\Delta n$ as a function of stable cavity radius for several different dislocation densities. Calculations for bridging a gap $r_c^c - r_c^s$ of 0.1 nm.
7. The ratio of $\sigma/\Delta n$ as a function of the difference $r_c^c - r_c^s$ for two dislocation densities and an assumed cavity radius, r_c^s, of 2 nm.
8. The required number of vacancies Δn as a function of the deficiency in gas atoms contained in a cavity, $\Delta n_g = n_g^* - n$.

9. Probability that a sequence of cascades provides Δn or more vacancies to a cavity as a function of Δn for various time intervals. The stepwise reduction arises because *the condition Δn or more* vacancies can be satisfied by unit numbers of cascades for a range of Δn.

10. As for Fig. 9 but for larger Δn and t. Stepwise variations at small values have been smoothed out.

11. Plot of Δn_g for which an arbitrarily chosen maximum value of Δn (1400) is sufficient to enable the cavity radius to increase from r^s to r_c by fluctuations. Also shown is n_g^* as a function of temperature.

12. Fractional change in dose to the onset of bias-driven swelling due to cascade-induced fluctuations, as a function of temperature. The two curves compare calculations where fluctuations are included and excluded.

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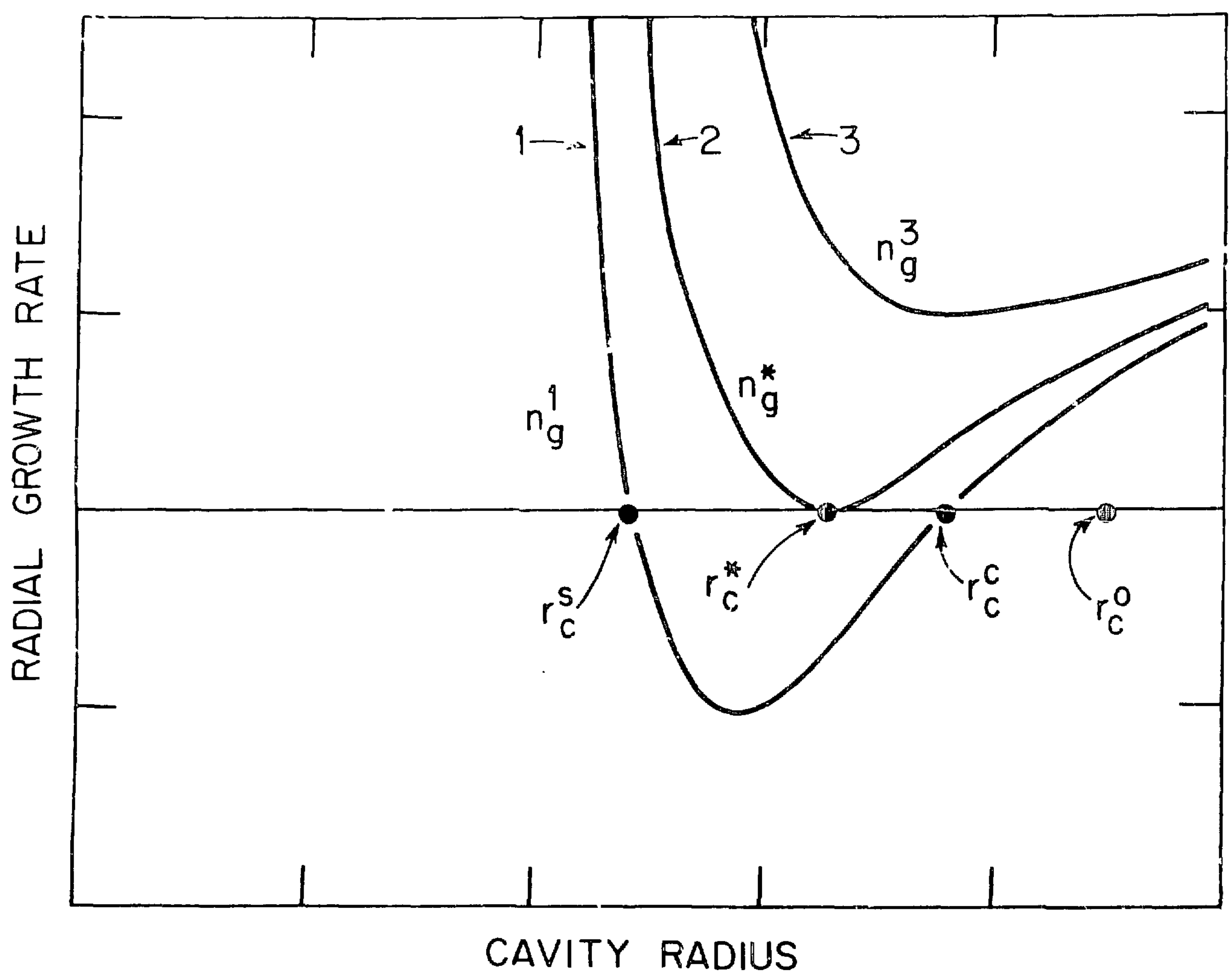

Fig 1, Hayns and Mansur

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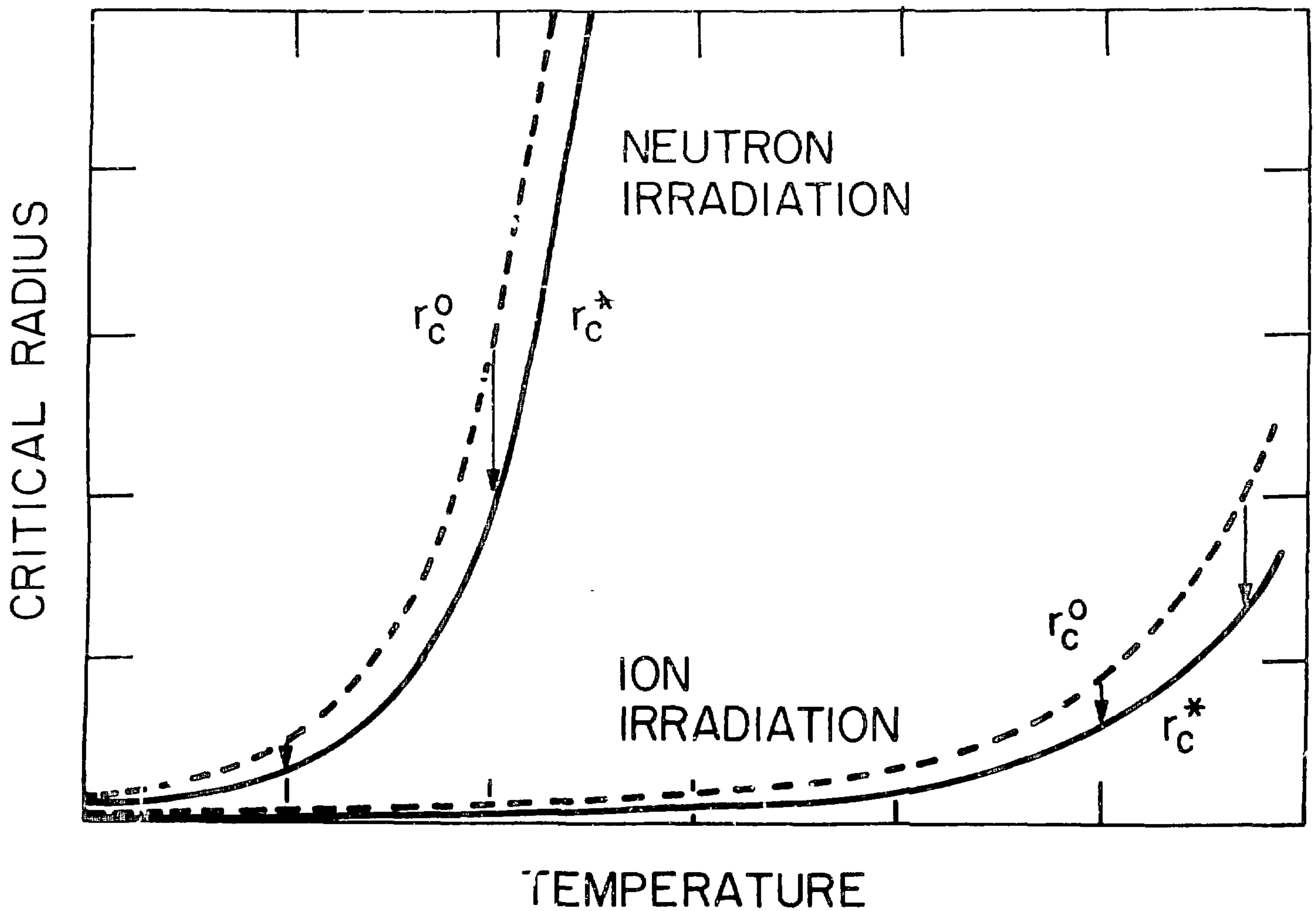

Fig 2, Heyns and Mansur

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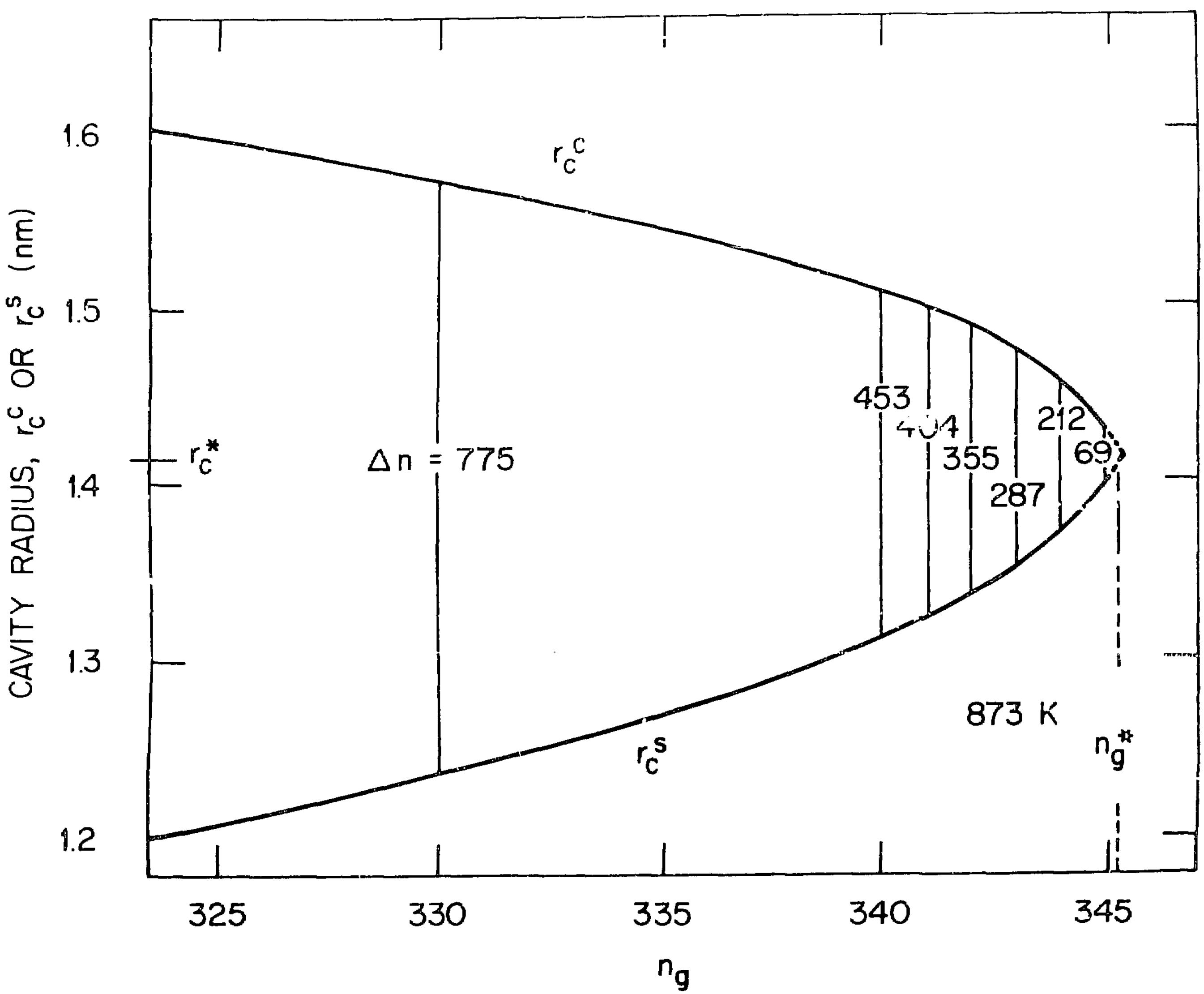

Fig 3, Hayns and Mansur

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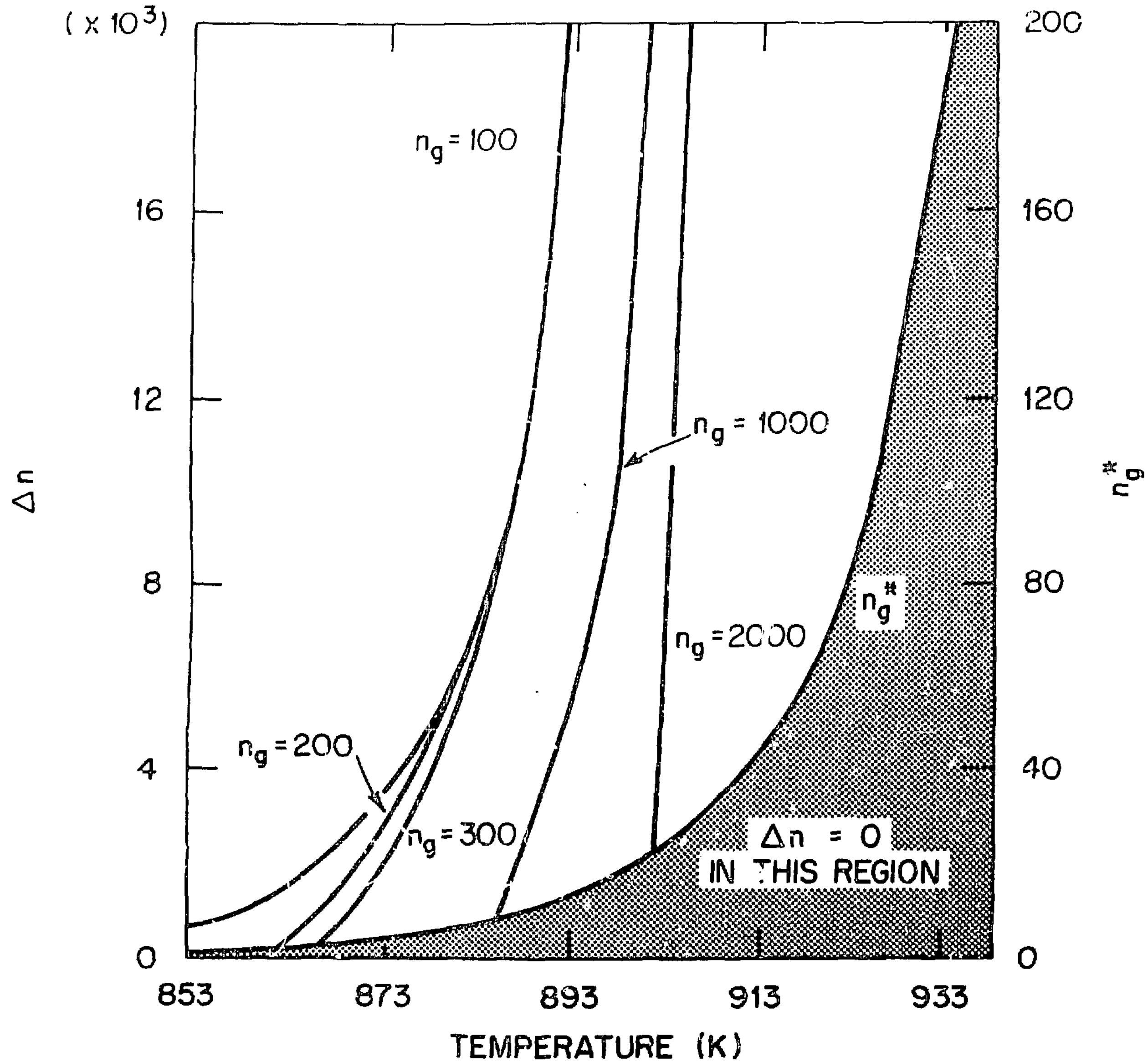

Fig 4, Haynes and Mansur

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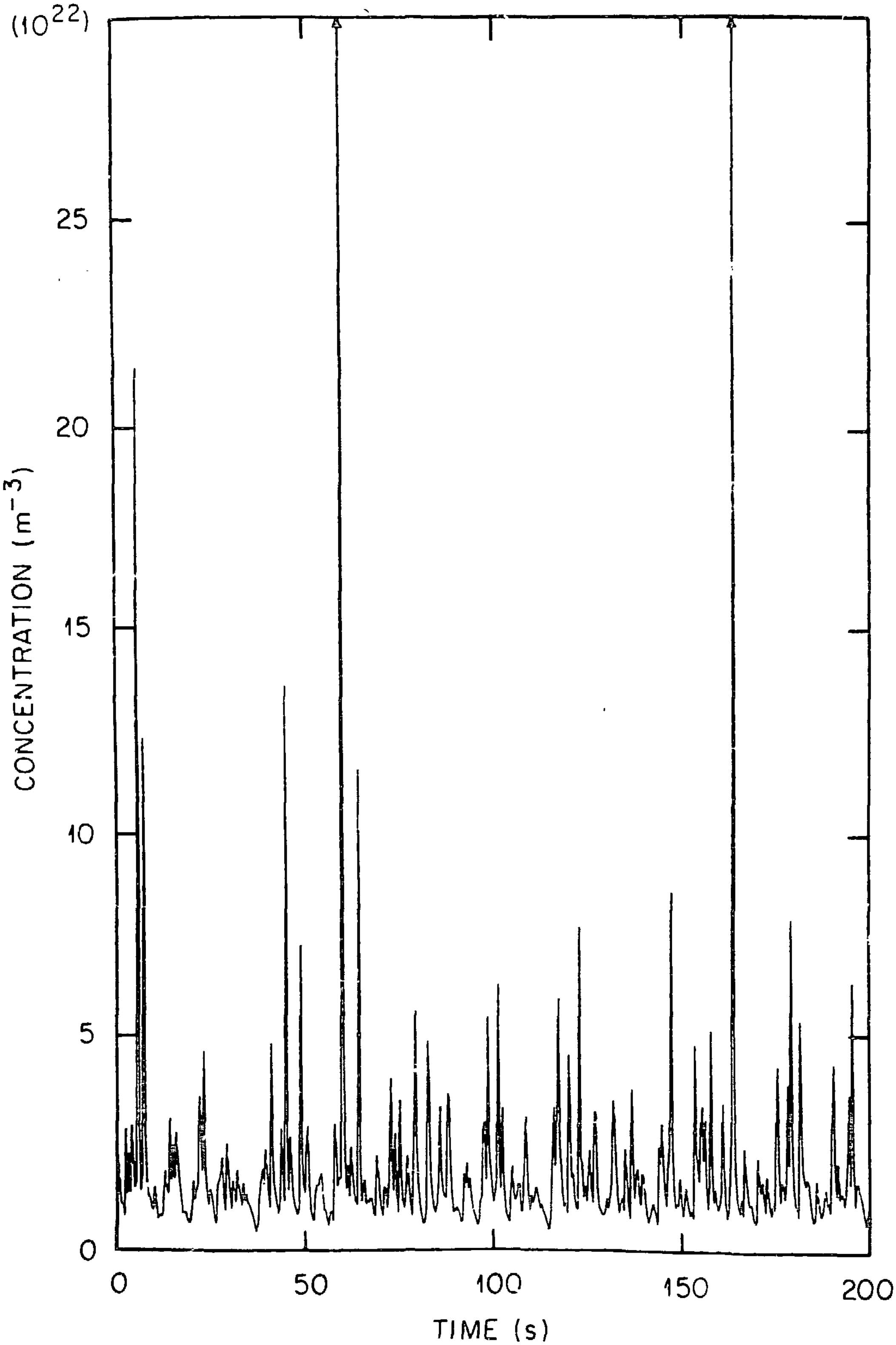

Fig 5, Hayes and Manner

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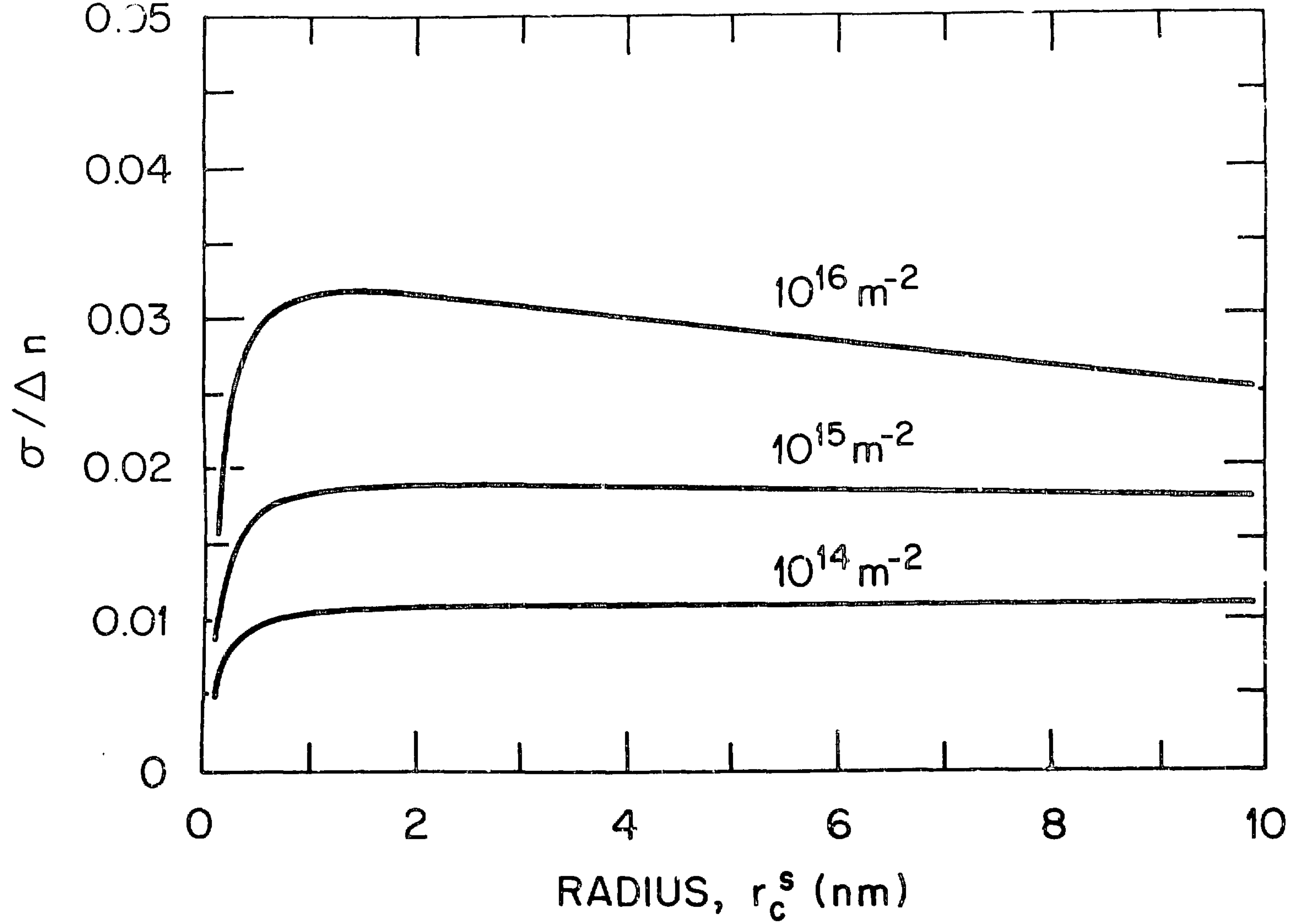

Fig 6, Haynes and Mansur

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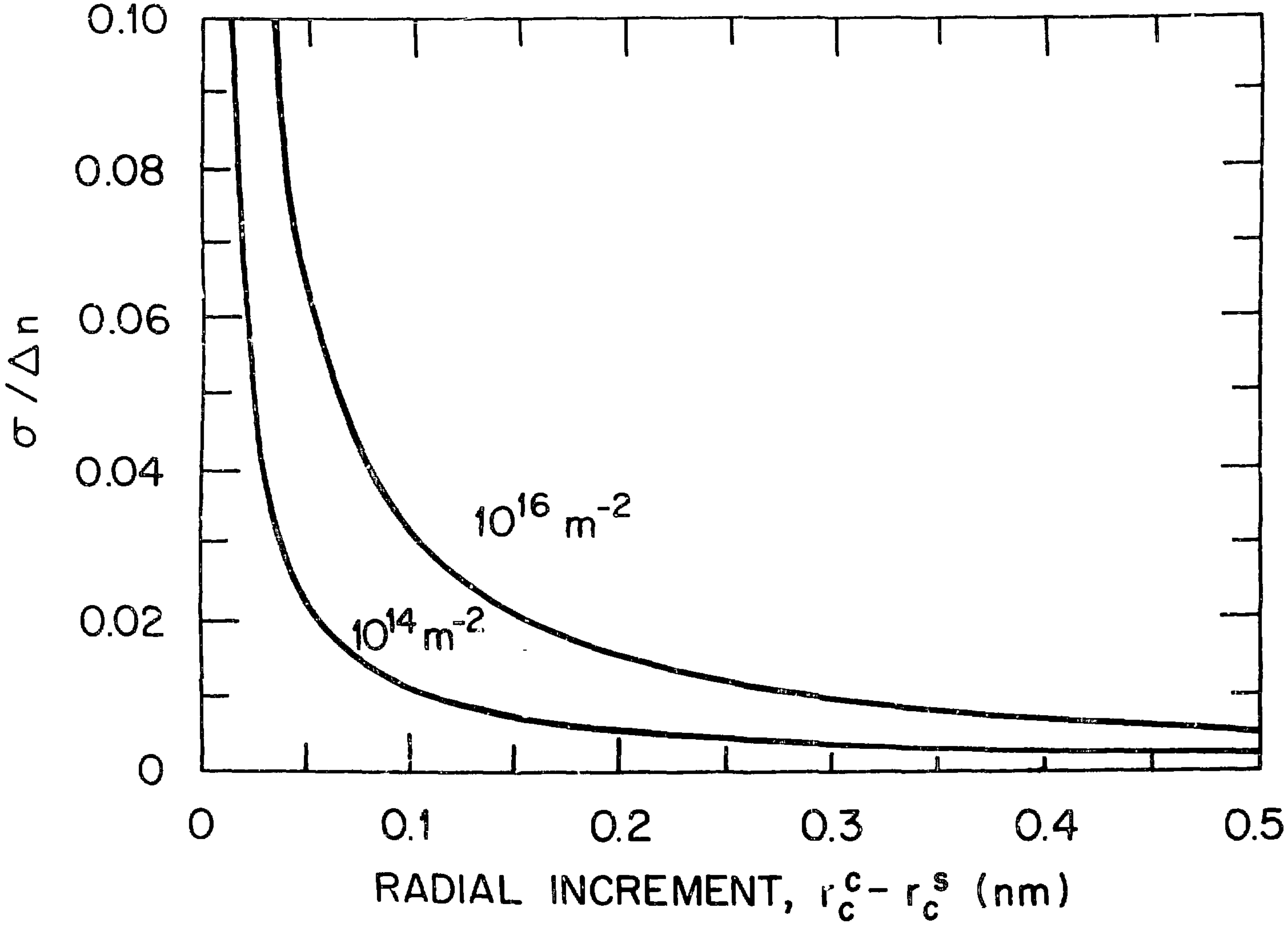

Fig 7, Haynes and Manner

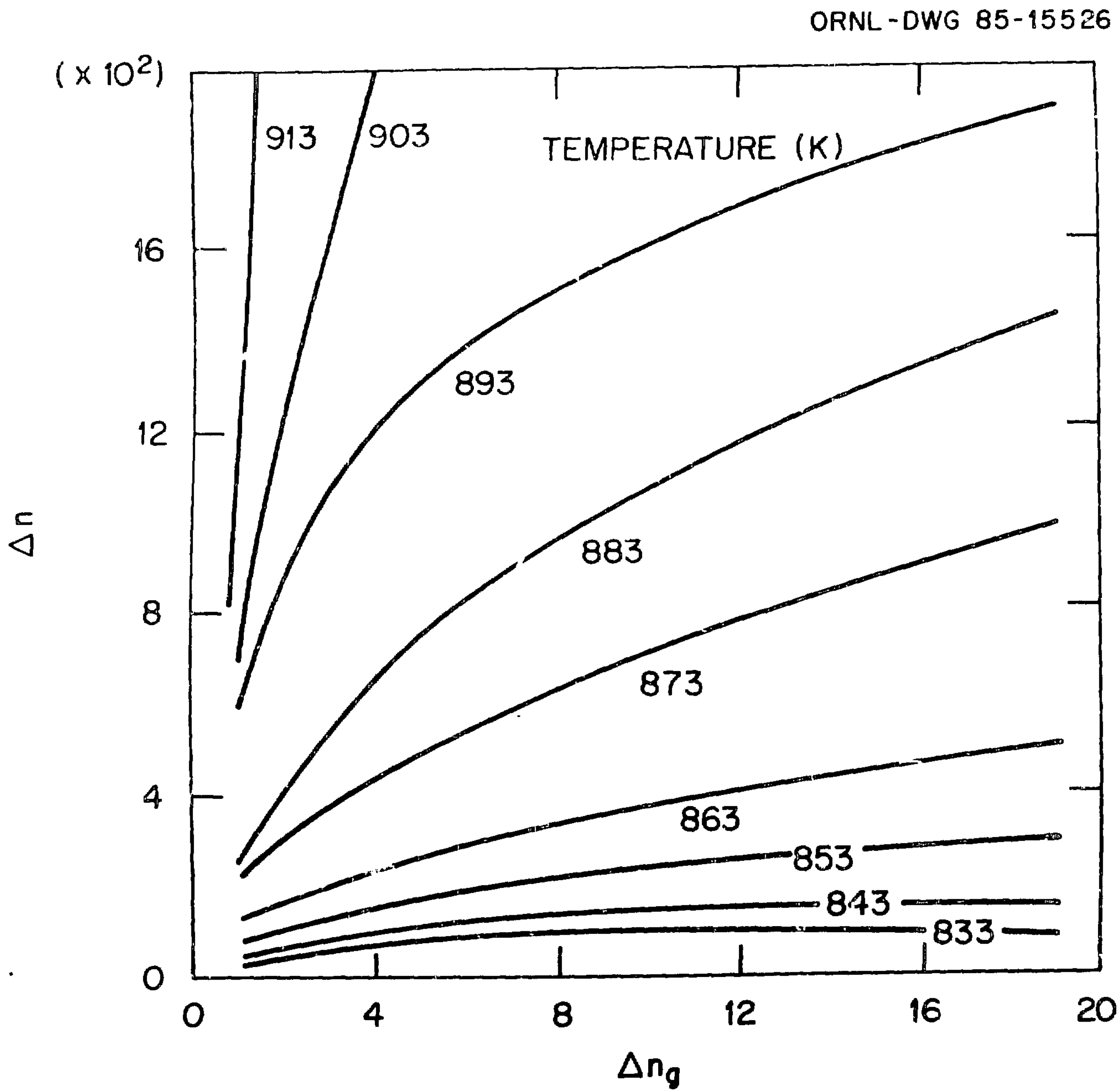

Fig 8, Haynes and Mansur

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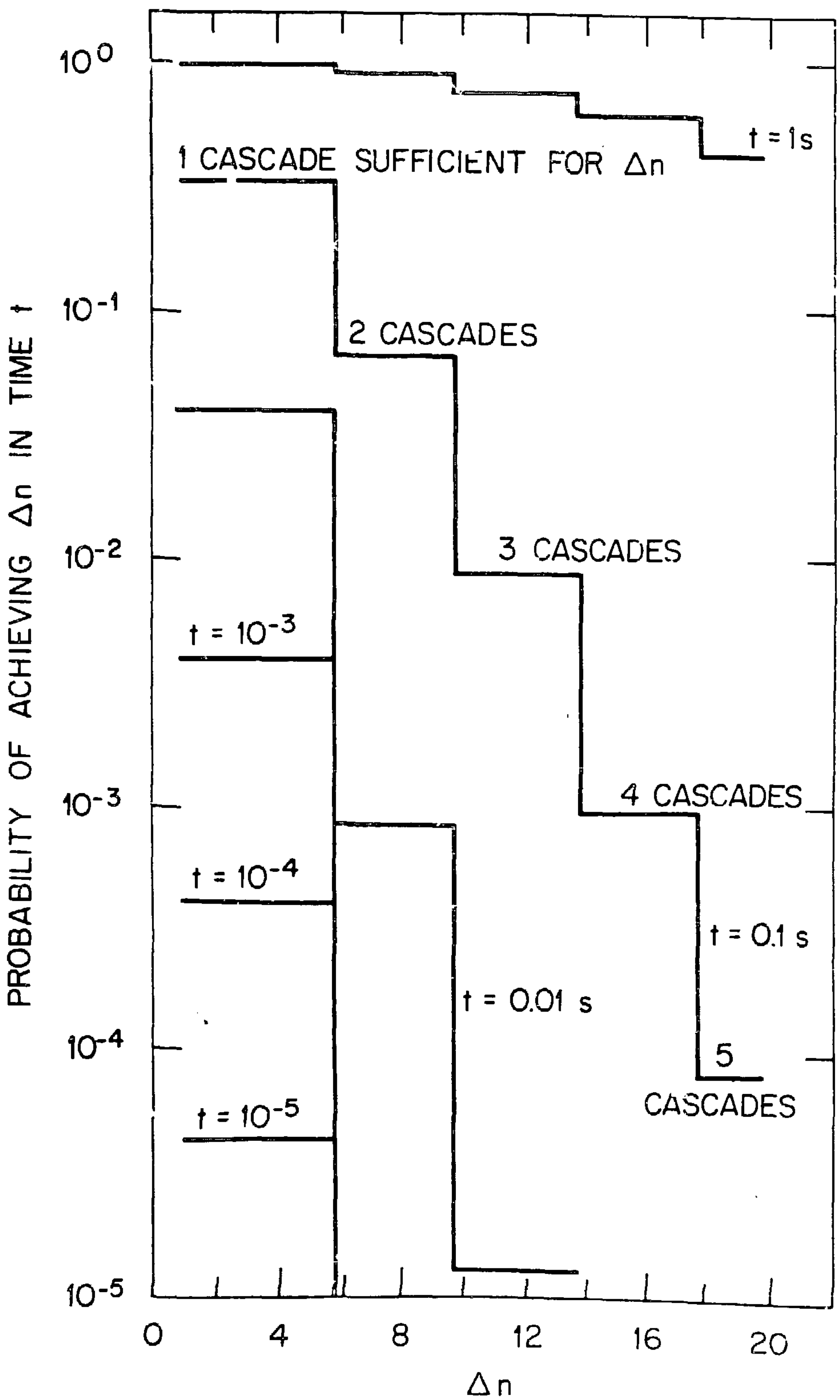

Fig 9, Hayns and Manson

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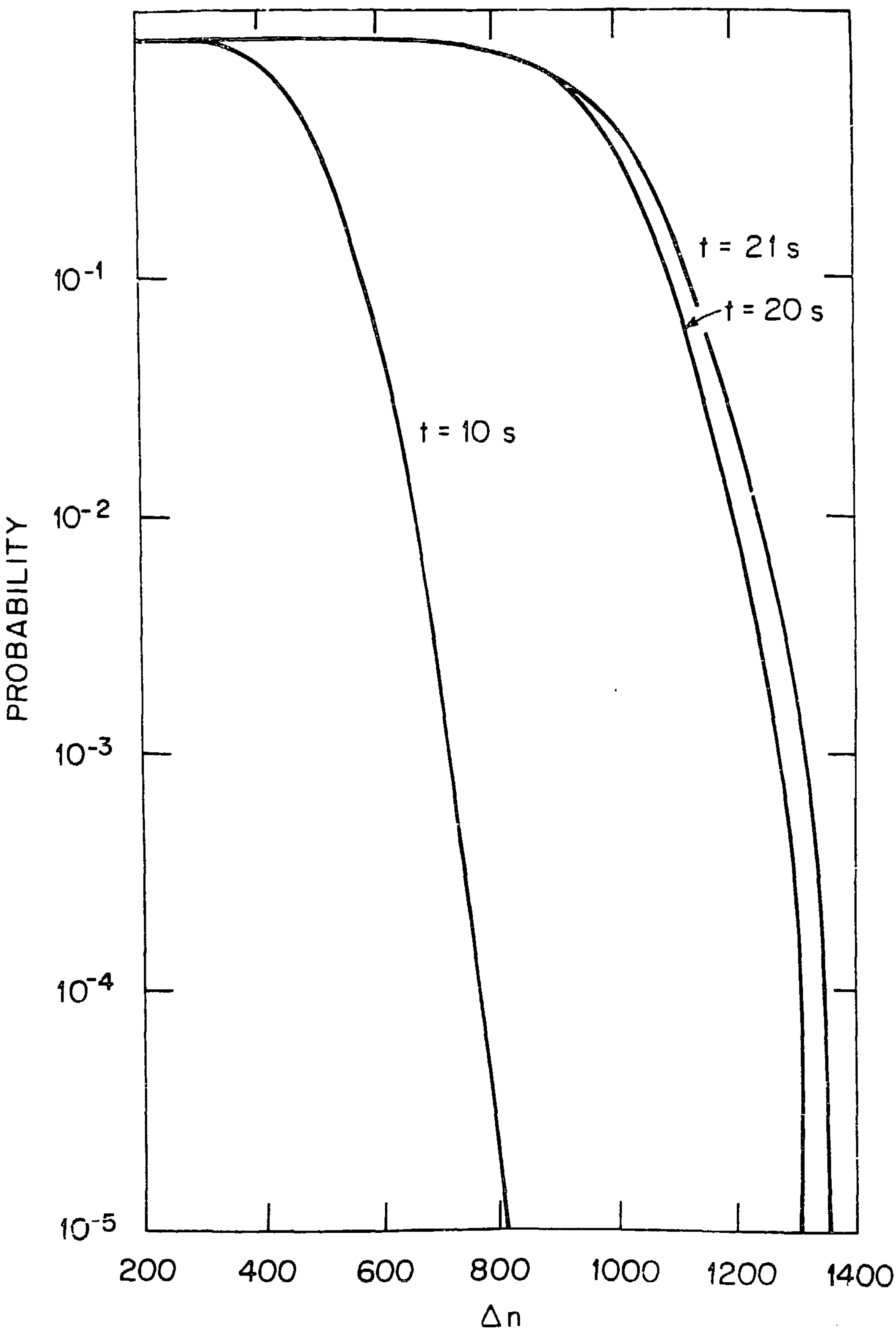

Fig 10, Hanns and Mavor

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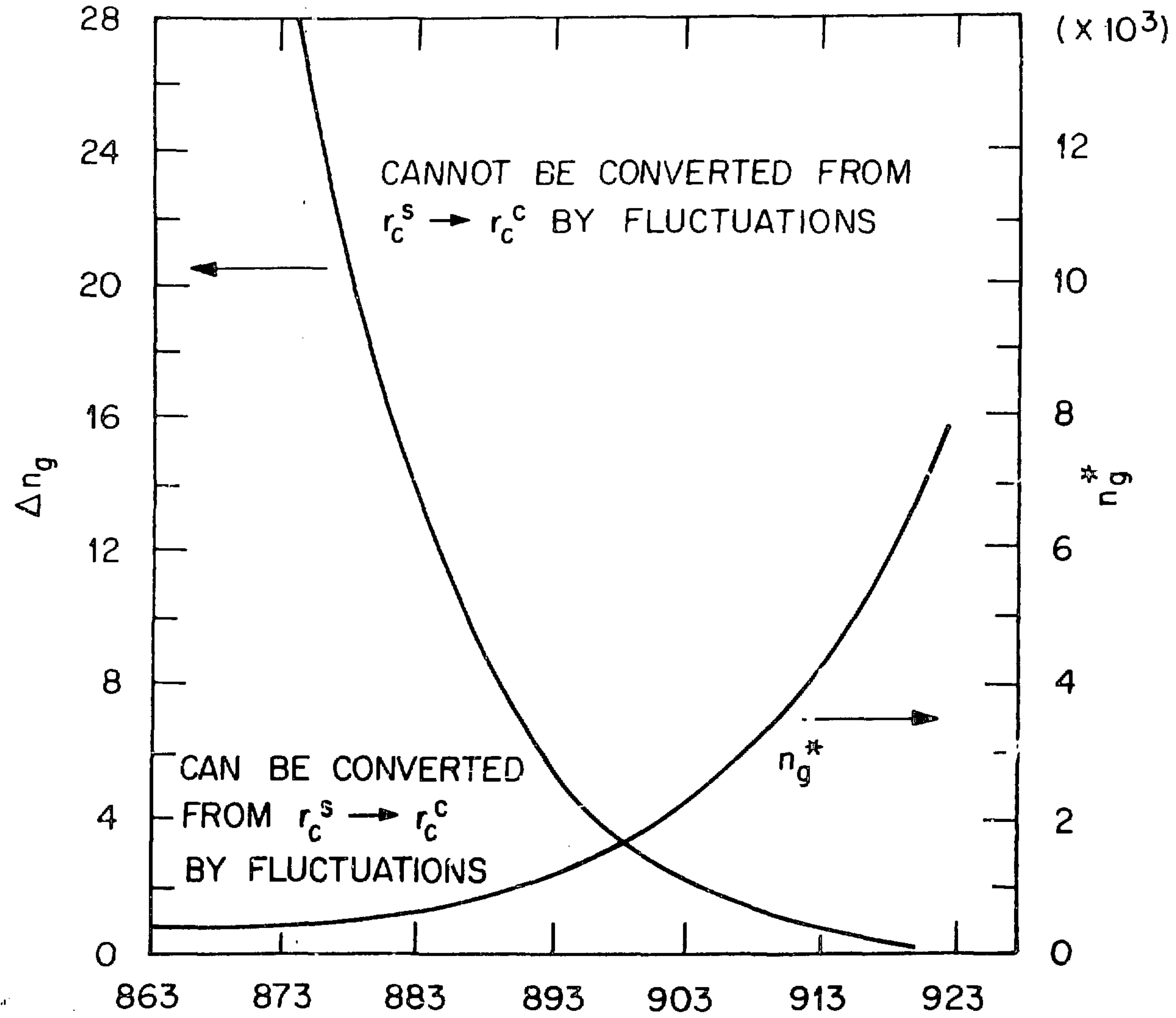

Fig 11, Haynes and Manner

ORNL-DWG 85-15522

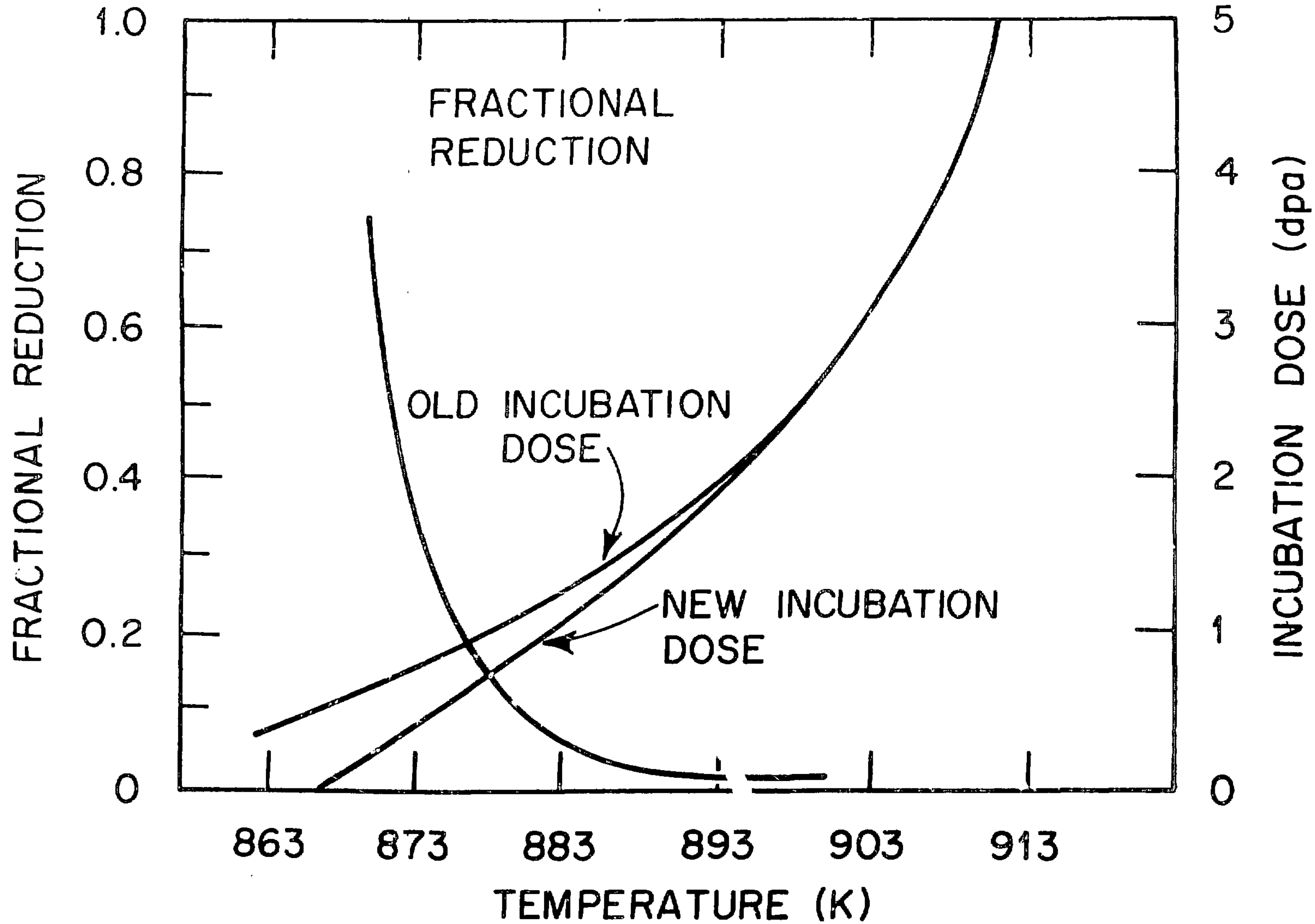

Fig 12, Haynes and Mansur