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BIAS FACTORS FOR RADIATION CREEP, GROWTH AND SWELLING

by

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ABSTRACT

Central to the present concepts of the origin of the radiation-induced creep, growth and swelling phenomena is the relative interaction of interstitials and vacancies with various sinks. Radiation-induced climb of dislocations, which figures in many theories of radiation creep and growth, requires the absorption of an excess of either vacancies or interstitials. On the other hand, radiation swelling requires the absorption of an excess of vacancies to effect void growth. These relative preferences are normally expressed

in theoretical models by certain bias factors, or capture efficiencies, usually assumed to be constant. Several attempts have been made to estimate their magnitude theoretically but all are seen to involve errors or physically unrealistic assumptions. We present here a unified treatment in which these various bias factors are estimated in a self-consistent model which incorporates, for the first time, all the essential physics, i.e., defect production, interactions of both vacancies and interstitials with sinks and the presence of two types of sinks. We present quantitative evaluations for the SIPA creep model and for radiation swelling, and compare with previous estimates of these phenomena.

1. Introduction

The physical origin of various phenomena occurring under irradiation, such as radiation growth, radiation creep and void swelling lies in the absorption of relatively more of one type of point defect at one type of sink (and consequently less of that same defect at another type of sink). The reason for such preferential absorption is generally considered to lie in the interaction energies between point defects and the various sinks.

The mechanisms of radiation growth are still not clearly established, but one contributing source, for cold-worked material at least, is probably the climb of dislocations having an anisotropic distribution of Burgers vectors[1]. It is considered that, due to its larger relaxation strain, the interstitial in metals interacts more strongly with a dislocation stress field than does the vacancy, and hence an excess of interstitials is absorbed by dislocations, thus causing their climb. Since in the steady state vacancies and interstitials must

disappear at exactly the rate at which they are produced, it is clear that another type of sink must exist which attracts interstitials less than do dislocations. These second sinks then absorb an excess of vacancies equal in magnitude to the excess of interstitials absorbed at dislocations. It is further clear that vacancy interactions with the different sinks produce analogous effects so that the overall net effect (or bias) is due to the combined, simultaneous interactive diffusion of both vacancies and interstitials to at least two types of sinks. The sinks having a smaller attraction for interstitials may be dislocation multipoles, dislocation cell walls, sub-boundaries, precipitates, voids or grain boundaries.

In radiation creep, two basically different processes have been considered. One is equivalent to that discussed under radiation growth but where the anisotropic distribution of Burgers vectors is no longer a requirement. In this concept, the dislocations climb does not itself produce creep strain but dislocation glide following the overcoming of some impedance actually produces the creep strain. Both internal stress fields[2] and local obstacles[3] have been considered by various authors. A second type of process has been proposed which requires climb alone to produce creep. This is the so-called SIPA (Stress Induced Preferential Absorption) model[4] and assumes that dislocations having different orientations of their Burgers vectors provide the required two types of sinks. Their asymmetric interactions with point defects are in turn supposed to arise from the shear polarizabilities of the defects[5]. The shear polarizability of a dumb-bell interstitial is assumed much larger than that of a vacancy and so the SIPA effect is felt to be due primarily to interstitials, with the vacancies partitioning themselves among the various dislocations in a relatively unbiased manner.

The void swelling phenomenon requires, obviously, a net absorption of vacancies by voids. This is generally thought to obtain primarily because of the preferential absorption of interstitials at dislocations, as discussed above. Thus dislocation climb (and possibly growth and/or creep) must accompany the growth of voids.

The "real" situation in which many sinks of various types (and geometries) are spatially distributed, each with its own diffusional fields of both vacancies and interstitials, has generally been considered too complex to analyze directly, even when defect-sink interactions are ignored. The model generally employed is one in which the concentration of each type of point defect is assumed constant throughout. The body and their losses to the various sinks as well as their annihilation by recombination are assumed to occur homogeneously. The rates at which these various losses occur must then be estimated by independent calculations. These involve the solution of a boundary-value diffusion problem where an individual sink of a particular type is represented by its actual geometry and size. Even when defect-sink interactions and recombination are ignored, various procedures have been employed to "couple" the discrete sink to the surrounding medium. Wiedersich[6] used the Wigner-Seitz cell approach and surrounded the sink with a sink-free region, within which point defects are generated, whose outer boundary was chosen to give the same cell volume as the average volume per sink in the actual medium and no flux was allowed to cross that outer boundary. In one approach, the discrete sink is surrounded with a "lossy medium", an infinite region in which defects are generated and are lost to other types of sinks at rates which vary spatially and are proportional to the local defect concentration. Brailsford and Bullough[7] placed a sink-free region between the discrete sink and the "lossy medium" but later Brailsford, Bullough and Hayns[8] removed it. All of these are approximations and it is difficult to select one appraoch over the other though it seems clearly best to

use an internally consistent set of such sink terms, derived by equivalent approaches. We recently suggested such a set.

When defect-sink interactions are included, the complexity increases significantly. Bullough, Eyre and Perrin[10] used a single cylindrical Wigner-Seitz cell surrounding a long dislocation line. Both vacancies and interstitials were assumed to be produced at equal rates and to disappear by recombination at a rate proportional to the product of their concentrations. The interstitials had a radially symmetric, attractive interaction with the dislocation and "zero-flux" boundary conditions were imposed for both defects at the outer cell boundary. As we discussed previously however[11], their use of only one type of sink in a region across whose boundary no defects pass requires that in the steady state both vacancies and interstitials enter the dislocation at precisely equal rates. Thus we attribute their finite bias factor to numerical errors, so that the bias factor of 2% which Brailsford and Bullough[7] later quote as being obtained from this model must be considered spurious.

Several authors[12,13] have employed a single diffusional cell around a dislocation and allowed not defect production. Defect production is simulated by assuming fixed defect concentrations at the outer boundary. With different interaction fields, then, for the same outer-boundary concentrations, vacancies and interstitials arrive at the central dislocation at different rates. This approach avoids the inconsistency of the Bullough, Eyre and Perrin appraoch but suffers from at least two crucial problems. First, the use of fixed concentrations (supplied by imaginary sources outside the diffusional field) introduces an unknown error. Second, its use of only one type of sink seems implausible for estimating an effect which physically demands at least two types of sink.

We previously discussed in some detail[11] the above approaches, along with others[12-19] used in estimating bias factors for both void swelling and SIPA-creep analyses, and concluded that none had quantitative reliability. We subsequently presented an analysis of the SIPA-creep process which included uniform defect production and the presence of both types of dislocation with their asymmetric interaction fields[20]. The resulting creep rate was shown to be significantly less than previous estimates. More recently, we have presented a simplified analysis for estimating the bias factor for void swelling[21]. We allowed both dislocations and voids but to simplify the geometry considered only cylindrical voids with a length per unit volume exactly equal to that of the dislocations. Interaction of interstitials with dislocations was allowed but, for simplicity, interactions of interstitials with voids and of vacancies with both dislocations and voids were ignored. The bias estimated was significantly larger than previous theoretical or "experimental" estimates.

Our purpose here is first to present a synopsis of the various types of "bias factors" introduced by different authors. Then we shall compare and contrast the various theoretical attempts at quantifying the bias. Then we shall present a unified analysis appraoch which treats defect production and allows for the coupled diffusional flow of defects in the presence of two types of sinks which interact differently with the point defects. Bias factors so derived will be compared with previous estimates. Finally, quantitative estimates of void swelling rates will be compared with experiment to bring into focus the need for the carefull assessment of <u>effective</u> defect production rate, i.e. the rate of production of freely mobile defects which escape annihilation within the damage cascade. This will also highlight the need for consistent

coupling between the bias factors and the various sink loss terms, both of which are treated simultaneously in the present approach, which also naturally supplies the volume-fraction and multiple-sink effects other authors have attempted to estimate by independent models.

2. Analysis

As has generally been done before, we shall ignore recombination in our model, so that recombination effects can only be assessed by assuming the appropriate defect production rate to be reduced by the recombination rate which in turn must be estimated independently. We shall also treat only two types of sinks, so that again the appropriate defect production rate must be reduced by the rate at which defects are independently estimated to be lost to other sinks. There is no obvious reason why additional types of sinks cannot be included directly in the model, however, and we expect to pursue this in future research.

With the above assumptions, conservation requires

$$\dot{a} = Z_{I1}\, I_1^o + Z_{I2}\, I_2^o = I_1^o + I_2^o \qquad (1)$$

and

$$\dot{a} = Z_{V1}\, I_1^o + Z_{V2}\, I_2^o = I_1^o + I_2^o \qquad (2)$$

where $\dot{a}$ is the effective defect production rate (assumed equal for vacancies and interstitials); Z_{I1}, Z_{I2} (Z_{V1}, Z_{V2}) are the ratios of loss rates of interstitials (vacancies) to sinks of types 1 and 2 with and without interaction

fields. The Z's so defined are equivalent to Mansur's sink capture efficiencies.

Equations (1) and (2) show clearly that the four Z's so defined are not independent. In fact, one easily sees that

$$Z_{I2} = 1 + \frac{I_1^o}{I_2^o} - Z_{I1}\frac{I_1^o}{I_2^o} \tag{3}$$

and

$$Z_{V2} = 1 + \frac{I_1^o}{I_2^o} - Z_{V1}\frac{I_1^o}{I_2^o} \tag{4}$$

where the I's are set by the densities and geometrical features of the sinks. Thus the Z's for each defect are related through the relative sink strengths. As shown in the reaction-rate-theory analysis by Wiedersich[6], the maximum in the rate of any process requiring the preferential absorption of one type defect at one sink occurs when the two sinks involved have equal strengths, i.e. when $I_1^o = I_2^o$. For this special case, (3) and (4) become

$$Z_{I2} = 2 - Z_{I1} \quad \text{(Equal sink strengths)} \tag{5}$$

and

$$Z_{V2} = 2 - Z_{V1} \quad \text{(Equal sink strengths)} \tag{6}$$

Since the limiting case is zero absorption at one type sink, say $Z_{I2} = 0$, it is clear that the maximum possible value of the Z for the other sink, say Z_{I1}, is 2, for this symmetrical case. The set $Z_{I1} = 2$, $Z_{I2} = 0$ expresses the physical situation when all the interstitials are absorbed at sinks of type 1 and none at sinks of type 2. Thus, the range of possible Z-values for this symmetrical

case is 0 to 2. On the other hand, when a large difference in sink strengths exists, say $I_1^o >> I_2^o$, Z_{I2} can become very large (small) when Z_{I1} is only very slightly less (greater) than unity. Of course, this simply reflects the physical fact that a small change in absorption at a sink of very high strength requires a proportionately large change in absorption at a weak sink.

It has been shown by rate-theory analyses assuming constant Z's that the rate of a process requiring differential absorption by one sink can be expressed

$$\text{Rate} \propto (Z_{I1} - Z_{V1}) - (Z_{I2} - Z_{V2}) \tag{7}$$

This combination of Z's is equivalent to Wiedersich's rate-theory expression for the rate of void swelling[6]

$$\text{Rate} \propto \beta - \alpha \tag{8}$$

when only dislocations and voids are present and where α and β describe the differencein capture efficiencies for interstitials and vacancies at voids and at dislocations, respectively, and are assumed to be constants. An alternative form of Eq. (7) is sometimes given

$$\text{Rate} \propto Z_{I1} Z_{V2} - Z_{I2} Z_{V1} \tag{9}$$

again assuming constant Z's.

The bias factors on the right-hand sides of Eqs. (7) and (9) can easily be shown to be equal by use of Eqs. (1) and (2). Thus, they may be considered

equivalent definitions of a bias factor for the particular rate process being analyzed.

Some authors attach a Z-factor to the term for interstitial loss-rate to dislocations and none to any other loss terms for either interstitials or vacancies. This is equivalent to assuming that all other Z's are unity, but as we saw above this is physically impossible. Such inconsistencies, however, can and do go completely unnoticed in a reaction-rate theory calculation where defect conservation is imposed on the system. And if the single Z-factor so assumed is treated strictly as an empirical factor with no physical meaning, well and good. However, it allows no judgments as to what a reasonable value would be. If such an assumption is inserted into Eqs. (7) or (9), the result is

$$\text{Rate} \propto Z - 1 \tag{10}$$

but such a treatment becomes inconsistent and unreliable when theoretical estimates of the effect of interaction fields on the defect fluxes into dislocations are used to obtain Z.

The various Z-factors discussed above, together with their corresponding sink strengths to which the I's of Eqs. (1) and (2) are proportional, embody most of the physics contained in reaction-rate-theory analyses of radiation-induced rate processes and we turn now to their evaluation.

a. SIPA Creep

We previously presented[20] an analysis of the SIPA creep model and will only give a summary here. In this case, the two types of sinks are long, parallel

dislocations whose Burgers vectors are such that an applied stress aids the absorption of one type of defect at one dislocation and not at the other. The interaction fields were those derived by Bullough and Willis[5], averaged over their attractive regions and applied as radially symmetric fields in the two cylindrical regions surrounding the two dislocations, each with a radius chosen so that the cell volume was equal to the average volume per unit length of dislocation in the real material. The concentrations of each type of defect were required to match at the outer boundary and the flux of each type of defect leaving one region was required to enter the other. The important interaction here is that due to the elastic polarizability of the point defect and we followed Bullough and Hayns[17] in neglecting the vacancy interactions. Thus, for this process,

$$\text{Rate} \propto Z_{I1} - Z_{I2} \tag{11}$$

where the Z's in this case are directly proportional to the applied stress. For values typical of stainless steels, we obtained bias factors which were weakly temperature dependent (especially in the range of usual interest) but somewhat dependent upon dislocation (sink) density. Typical results are shown in Fig. 1. The curve labeled "simple" assumed interstitials in each cylindrical region interact only with that dislocation, whereas the curve labeled "compound" assumed that in each region the interaction energies of the two neighboring regions are linearly additive. The effect of this sink "competition" is relatively minor even at very large dislocation densities. For an applied tensile stress of 100 MPa and a dislocation density of 6×10^{14} m/m^3, a bias factor

$(Z_{I1} - Z_{I2}) \sim 0.08\%$ was obtained and the predicted creep rates, as shown in the figure, were considerably lower than the rate previously predicted by Bullough and Hayns[17].

b. Dislocation - Cylindrical Void

We recently presented[21] a model much like the SIPA model above but one in which the econd cylindrical region contains not a dislocation but a cylindrical void. Here the major source of the interactions is the relaxation strain of a point defect. As a first approximation, we neglected interstitial interactions with the void and again neglected all vacancy interactions. Only one temperature and one dislocation density were considered and the results for Z_{ID}, the capture efficiency of interstitials (assuming a relaxation strain of unity) at dislocations, is shown in Fig. 2 as a function of void size. The corresponding values of Z_{IC}, the capture efficiency of interstitials at cavities (voids) were not reported but have been obtained here from Eq. (3) and are also plotted. With these two Z's, the void-swelling bias factor was also calculated from Eq. (11) and plotted in Fig. 2. Thus, for equal sink strengths of dislocations and cylindrical voids, this simple model predicts a bias factor $Z_{ID} - Z_{IC} = 1.14$ and, for a void sink strength greatly exceeding the dislocation sink strengths, values approaching 2 are predicted. This range corresponds to a range in a single effective Z using Eq. (10) from $Z \sim 2.1$ to $Z \sim 3$. These are much higher

than most authors assume in their reaction-rate-theory models, but we defer any more quantitative comparisons to the new model to be presented below which will utilize interactions of both interstitials and vacancies with both dislocations and spherical voids where both sinks may be present in arbitrary densities.

c. Dislocations and Spherical Voids

Consider a cylindrical region, of outer radius R, containing a long, straight dislocation, of core radius r_o, at its center. Ignoring recombination, we can write, for either type of defect, the general solution to the Poisson equation for cylindrical symmetry

$$J = \frac{\mathring{a}r}{2} + \frac{b}{r} \tag{12}$$

where J is the defect flux, $\mathring{a}$ is the defect production rate (assumed equal for vacancies and interstitials) and b is a constant. We also assume the usual flux equation

$$J = - \frac{D'c}{kT} \nabla\mu \tag{13}$$

where D' is the local defect diffusivity (as affected perhaps by interaction fields), c is the defect concentration, μ is its chemical potential, k is Boltzmann's constant and T is absolute temperature. We assume the usual Arrhenius relationship for D'

$$D' = De^{-\left(\frac{E^S - E^G}{kT}\right)} \tag{14}$$

and express the chemical potential as

$$\mu = kT \ln c/c_{eq} + E^G \tag{15}$$

where D is the defect diffusivity in the absence of interaction fields, E^S and E^G are its interaction energy in the saddle-point and ground-state, respectively, and C_{eq} is the equilibrium defect concentration. Inserting Eqs. (14) and (15) into (13) gives

$$J = - De^{-\left(\frac{E^S - E^G}{kT}\right)}\left[\nabla c + \frac{c}{kT} \nabla E^G\right] \tag{16}$$

which is easily rewritten in the form

$$J = - De^{- E^S/kT} \nabla \left(ce^{E^G/kT}\right) . \tag{17}$$

For the case of radial symmetry, Eq. (17) becomes

$$J = - De^{-E^S/kT} \frac{d}{dr}\left(ce^{E^G/kT}\right) . \tag{18}$$

Equating (18) and (12) and integrating, we obtain

$$b_D = - \frac{D\left[c\, e^{E_D^G/kT}\right)_R - ce^{E_D^G/kT}\Big)_{r_o}\Big]}{\int_{r_o}^{R} \frac{e^{E_D^S/kT}}{r}\, dr}$$

$$- \frac{\dot{a}\int_{r_o}^{R} re^{E_D^S/kT}\,dr}{2\int_{r_o}^{R} \frac{e^{E_D^S/kT}}{r}\,dr} \tag{19}$$

where the subscript "D" indicates the dislocation cell.

Now, consider a spherical region with the same outer radius R, containing at its center a void of radius r_v. Assuming spherical symmetry, the solution to Poisson's equation is

$$J = \frac{\dot{a}r}{3} + \frac{b}{r^2} \tag{20}$$

which in turn is set equal to Eq. (18) to obtain, after integrating,

$$b_C = - \frac{D\left[\left(ce^{E_C^G/kT}\right)_R - \left(ce^{E_C^G/kT}\right)_{r_v}\right]}{\int_{r_v}^{R} \frac{e^{E_C^S/kT}}{r^2}\,dr}$$

$$- \frac{\dot{a}\int_{r_v}^{R} re^{E_C^S/kT}\,dr}{3\int_{r_v}^{R} \frac{e^{E_C^S/kT}}{r^2}\,dr} \tag{21}$$

where the subscript "C" signifies the void (cavity) cell. Since we are omitting recombination, vacancies and interstitials are independent and there are a total of four "b" constants, one for each type of defect in each cell, and of course four E functions as well.

For boundary conditions, we first take

$$ce^{E_D^G/kT}\Big)_R = ce^{E_C^G/kT}\Big)_R \qquad (22)$$

which represents two conditions, one for each type of defect. For all interactions of interest $E^G \to 0$ for large r, so that Eqs. (22) basically express continuity in defect concentrations at the outer boundaries of the two types of regions. One may of course add the interaction fields of neighboring cells so that $E_D^G = D_C^G$ is guaranteed identically but explicit calculations in the simpler models discussed in a. and b. above showed that such a refinement causes only small changes (less than a few per cent) in calculated results for any reasonable range of parameters. Therefore, we shall use here, for each cell, the interaction fields which would exist in an infinite medium containing only that sink.

For our third and fourth boundary conditions, we take

$$2\pi R\rho J_D(R) = -4\pi R^2 N_v J_C(R) \qquad (23)$$

for each type of defect, where ρ is the dislocation line length, and N_v is the number of voids (both per unit volume of solid).

Eq. (23) may be derived by writing the two conservation equations (for each type of defect) for the two cells

$$\dot{a} \cdot \pi (R^2 - r_o^2)\rho = - J_D(r_o) \cdot 2\pi r_o \rho + J_D(R) \cdot 2\pi R\rho \tag{24}$$

and $$\dot{a} \cdot \frac{4}{3}\pi(R^3 - r_v^3) N_v = -J_c(r_v) \cdot 4\pi r_v^2 N_v + J_c(R) \cdot 4\pi R^2 N_v \tag{25}$$

which simply state that all defects generated in each cell per unit time must, in the steady state, either enter the sink in that cell or diffuse out of the cell. Adding these together, we obtain

$$\dot{a} = - J_D(r_o) \cdot 2\pi r_o \rho - J_c(r_v) \cdot 4\pi r_v^2 N_v$$

$$+ J_D(R) \cdot 2\pi R\rho + J_c(R) \cdot 4\pi R^2 N_v \tag{26}$$

where we have used the identity

$$\pi(R^2 - r_o^2)\rho + \frac{4}{3}\pi(R^3 - r_v^3)N_v = 1 \tag{27}$$

which forms our <u>definition</u> of the outer radius R of the two types of cells. But since physically all defects must be absorbed at either the dislocations or the voids, we also have

$$\dot{a} = - J_D(r_o) \cdot 2\pi r_o \rho - J_c(r_v) \cdot 4\pi r_v^2 N_v \tag{28}$$

Equating Eqs. (26) and (28) leads directly to our boundary condition (23), which clearly is valid also for thermal fluxes which must originate at one type of sink and be absorbed at the other.

The boundary conditions (22) and (23) enable us to evaluate b_D and b_C and so obtain the fluxes

$$J_D = \frac{2\frac{N_v}{\rho} D \left[ce^{E_D^G/kT} \right)_{r_o} - ce^{E_C^G/kT} \left)_{r_v} \right]}{r\left[\frac{2N_v}{\rho} \int_{r_o}^{R} \frac{e^{E_D^S/kT}}{r} dr + \int_{r_v}^{R} \frac{e^{E_C^S/kT}}{r^2} dr \right]}$$

$$+ \frac{\dot{a}r}{2} - \frac{\dot{a}\left(\frac{2}{3} \frac{N_v}{\rho} R^3 + \frac{R^2}{2}\right) \int_{r_v}^{R} \frac{e^{E_C^S/kT}}{r^2} dr}{r\left[\frac{2N_v}{\rho} \int_{r_o}^{R} \frac{e^{E_D^S/kT}}{r} dr + \int_{r_v}^{R} \frac{e^{E_C^S/kT}}{r^2} dr \right]}$$

$$- \frac{2\frac{N_v}{\rho} \dot{a} \left[\frac{1}{2} \int_{r_o}^{R} re^{E_D^S/kT} dr - \frac{1}{3} \int_{r_v}^{R} re^{E_C^S/kT} dr \right]}{r\left[\frac{2N_v}{\rho} \int_{r_o}^{R} \frac{e^{E_D^S/kT}}{r} dr + \int_{r_v}^{R} \frac{e^{E_C^S/kT}}{r^2} dr \right]} \qquad (29)$$

and $$J_C = - \frac{D \left[ce^{E_D^G/kT} \right)_{r_o} - ce^{E_C^G/kT} \left)_{r_v} \right]}{r^2 \left[\frac{N_v}{2\rho} \int_{r_o}^{R} \frac{e^{E_D^S/kT}}{r} dr + \int_{r_v}^{R} \frac{e^{E_C^S/kT}}{r^2} dr \right]}$$

$$+ \frac{\dot{a}r}{3} - \frac{\dot{a}\left(\frac{2}{3}\frac{N_v}{\rho}R^3 + \frac{R^2}{2}\right)\int_{r_o}^{R}\frac{e^{E_D^S/kT}}{r}dr}{r^2\left[\frac{N_v}{2\rho}\int_{r_o}^{R}\frac{e^{E_D^S/kT}}{r}dr + \int_{r_v}^{R}\frac{e^{E_C^S/kT}}{r^2}dr\right]}$$

$$+ \frac{\dot{a}\left[\frac{1}{2}\int_{r_o}^{R} re^{E_D^S/kT}dr - \frac{1}{3}\int_{r_v}^{R} re^{E_C^S/kT}dr\right]}{r^2\left[\frac{N_v}{2\rho}\int_{r_o}^{R}\frac{e^{E_D^S/kT}}{r}dr + \int_{r_v}^{R}\frac{e^{E_C^S/kT}}{r^2}dr\right]} \qquad (30)$$

for each defect in each cell.

The Z-factors are obtained from

$$Z_{ID} = \left.\frac{J_{ID}(E)}{J_{ID}(0)}\right|_{r_o} \qquad (31)$$

and analogous equations for Z_{IC}, Z_{VD} and Z_{VC}, where the flux ratios are defined with all E's present in the numerator and all E's = 0 in the denominator.

Given the J's from Eqs. (29) and (30) we can find the swelling rate

$$\delta \equiv \frac{d}{dt}\left(\frac{\Delta v}{v}\right) = 4\pi r_v^2 N_v \left[J_{IC}(r_v) - J_{VC}(r_v)\right] \qquad (32)$$

where we have defined swelling as the increase in volume per unit volume of <u>solid</u>, not per unit <u>total</u> volume.

Similarly, the average climb velocity of the dislocations is given by

$$V_D = 2\pi \left[J_{VD}(r_o) - J_{ID}(r_o) \right] \tag{33}$$

where we have taken positive climb to correspond to interstitial absorption, and have taken r_o to be equal to the Burgers vector.

To evaluate Eqs. (29) and (30) we require values for the interaction energies. Here we use the analysis of Bullough and Willis[5] but omit the bulk modulus and shear modulus terms because they are only small fractions of the "misfit" terms given by

$$E_D^G = E_D^S = - \frac{r_o \Omega \mu \left| e^\circ \right|}{r} \tag{34}$$

where Ω is the atomic volume, μ is the shear modulus, e° is the point defect relaxation strain; we have assumed the interactions in the ground and saddle-point configurations to be essentially the same and have taken the average value over the attractive portion of the field. The first assumption is partly justified by the experimental observation[23] that the activation volume for interstitial motion in tungsten is very small. Activation volumes for vacancy migration are generally considered to be quite small also[24]. The second assumption is known to give very accurate results for the case of diffusion in a dislocation field without defect production[25].

For the interaction energies between point defects and voids, we use the misfit term of the analysis of Wolfer and Ashkin[16]

$$E_C^G = E_C^S = -\frac{\Omega^2 (e^\circ)^2 \mu(1+\nu)^2}{36\pi (1-\nu) r_v^3} \cdot \frac{2}{\xi^4(\xi+1)} \left[\frac{\xi^3}{(\xi-1)^3} - \frac{\xi^2+1}{\xi^2-1}\right] \quad (35)$$

where ν is Poisson's ratio and $\xi = r/r_v$. Again we have assumed equality of the interactions in the ground and saddle-point configurations.

3. Results

Before presenting numerical results, it is useful to point out some general characteristics of Eqs. (29) and (30), some of which also appeared in the results for SIPA creep and for the dislocation-cylindrical void problem. The first terms of the equations give the thermal contribution to mass-transport, they are directly proportional to the defect diffusivity and are non-zero only when the product of c and $e^{E^G/kT}$ differ at the two sinks. If local equilibrium is assumed,

$$c = c^\circ e^{-E^G/kT} \quad (36)$$

at each sink and the interaction energy disappears. The concentration c° may then be written

$$c^\bullet = c_{eq}\, e^{\frac{\Delta W}{kt}} \quad (37)$$

where ΔW is the reversible work done in the transfer of a defect across a sink interface (opposite signs for vacancies and interstitials). If there is an externally applied hydrostatic pressure, p, and a gas is present in the

void at a pressure, p_g, then we have

$$\Delta W_D = \pm p\Omega \tag{38}$$

and

$$\Delta W_C = \pm \left(p_g - \frac{2\gamma}{r_v} \right) \tag{39}$$

at the dislocation core and void, respectively, where γ is the surface tension. The assumption of local equilibrium is not essential but gives the maximum possible, "diffusion-controlled" rate. Alternatively, concentrations differing from local equilibrium might be assumed to simulate "interface control". Under thermal conditions ($\dot{a} = 0$) the current of each type of defect leaving one type of sink is exactly equal to the current entering the other, as holds for all mass-transport processes. This thermal swelling, or sintering depending upon the relative magnitudes of Eqs. (38) and (39), in principle includes contributions from both vacancies and interstitials but, for metals, the equilibrium value of interstitial concentration is usually assumed to be so small that primarily vacancies contribute. Finally, we note that the interaction energies in the saddle-point affect the thermal rate through the integrals in the denominators, and thus Z's (thermal) $\neq 1$ but they are <u>not</u> equal to the Z's defined above for the radiation-induced components.

The radiation-induced portions of the fluxes are seen to be independent of the defect diffusivities and directly proportional to the defect production rate. They are also <u>totally independent</u> of the boundary conditions at the two types of sinks. Hence, they are the same whether "diffusion control" (local equilibrium

concentrations at sinks) or "interface control" (sink concentrations fixed at non-equilibrium values) is assumed to apply. Thus, we question the validity of the distinction some authors have claimed[22,26,27] for these two cases using reaction-rate theory with the same Z's applied to both the radiation-produced defect absorption terms and the so-called thermal-emission terms. It is clear from Eqs. (29) and (30) that the interaction fields do not affect thermal fluxes in the same manner in which they affect the radiation-induced components, so that using the same Z's is inappropriate. This illustrates one of the difficulties in employing an approach such as reaction-rate theory where sink-loss terms and bias factors must be supplied from independent (and not necessarily wholly consistent) models. In the present treatment, no such externally calculated parameters are required since they are inherent in the model.

In Fig. 3 we show illustrative results for predicted swelling rates as a function of temperature. We have chosen our "standard" parameters to be representative of stainless steel, with[22] $D_I C_I = 1.58\times10^{-8}\ \exp(-51{,}000/T)\ m^2\cdot s^{-1}$, $D_V C_V = 10^{-5}\ \exp(-33{,}950/T)\ m^2\cdot s^{-1}$, $r_o = 0.126$nm, $\Omega = 1.20\times10^{-29}\ m^{-3}$, $\mu = 8.6\times10^4$ MPa, $e_I^o = 1.4$[28], $e_V^o = -0.23$[28], $\nu = 0.3$, $\gamma = 1.5\ J\cdot m^{-2}$. All of the cases shown in Fig. 3 employ $\rho = 6\times10^{14}\ m/m^3$, the single lower curve with a very weak cavity sink strength ($r_v = 10r_o$, $N_v = 10^{19}\ m^{-3}$) and $\dot{a} = 10^{-7}$ dpa $\cdot\ s^{-1}$ and the upper family of curves with a relatively high cavity sink strength ($r_v = 100r_o$, $N_v = 10^{21}\ m^{-3}$) and $\dot{a} = 10^{-6}$, 10^{-4} and 10^{-2} dpa $\cdot\ s^{-1}$. The radiation-induced swelling rate is seen to increase by about a factor of two from T = 100 to the maximum where thermally-induced cavity shrinkage causes a sharp decline. The

peak swelling in the upper family of curves is higher by over four orders-of-magnitude even though the defect production rate is only one order-of-magnitude higher. This reflects the fact that in the second case the two sink strengths are comparable, whereas in the first case they are widely different. As shown in the analysis of Wiedersich[6], maximum swelling rate obtains for equal sink strengths. This much higher swelling rate causes the peak to occur at significantly higher temperature. Since each case is for fixed sink strengths, it is clear that the bias factor for swelling is somewhat temperature dependent, though for the temperature range of significant swelling the variation is quite limited. We point out that each of these curves assumes a constant "effective" å, i.e. a constant rate of formation of defects which are absorbed at the two types of sinks considered. If å is interpreted as the actual defect product rate minus those absorbed at any other sinks, these curves still require multiplication by the S -parameter defined by Wiedersich[6] as the fraction of defects which are in fact absorbed at sinks. Such a factor would lower the low-temperature portions of these curves much more abruptly as temperature decreases and recombination removes essentially all defects. We estimate from Widersich's curves that $S \lesssim 10^{-2}$ below ~ 600-700K. Thus, the temperature variation of bias is indeed minimal within the swelling regime.

The increase of the temperature of peak swelling rate as å increases, along with a broadening of the temperature range of significant swelling, is clearly displayed by the upper family of curves in Fig. 3. The peak swelling rate is predicted to occur for the assumed sink structure at ~ 800, 1100 and 1300K for $å = 10^{-6}$, 10^{-4} and 10^{-2}, respectively. These three production rates are generally

considered to be roughly representative of fast reactor, fast ion and electron bombardment, respectively.

A few calculations were run under the conditions of the upper family of curves, but for $\dot{a} = 10^{-6}$ only, to assess the effects of an externally applied hydrostatic tensile stress and a gas in the cavities. The dotted curves show the trend for externally applied stress with the numbers on the curves indicating the magnitude of negative pressure (in MPa) applied. At 600K, stresses of ~0.1μ are required for any discernible effect; at 900K, stresses in excess of ~ 10^{-3}μ are required. The possibility of loading a metal to such high levels of hydrostatic tension without yielding seems marginal, so that for these conditions stress-enhanced swelling is unlikely to be measurable. Gas pressures, p_g, of comparable levels are required for significant effects on swelling rate, as indicated in the figure.

The general effect of variations in relative sink strengths at T = 600K and $\dot{a} = 10^{-6}$ for the combinations run to date is displayed in Fig. 4. The peaking when void and dislocation sink strengths are comparable is clearly displayed using the simple sink strengths normally employed. In some calculations N_v was varied, in others ρ and in still others r_v. However, very little overlap in relative sink strengths existed for the different sets of calculations. We shall discuss below a more precise function for correlating with sink strengths evaluated internally from the present model.

Fig. 5 displays the effect of variations in e_I^o and e_V^o, the relaxation strains of the point defects. It is of course the relative values of these parameters through their proportional effects on the interaction energies which principally

give rise to the void swelling phenomenon so the cross-over from shrinkage to swelling occurs of course precisely where e_I° exceeds e_V°. (Variations in e_V° were run with e_I° at its "standard" value, and vice versa.) The far-from-linear dependence reflects the fact that the fluxes involve integrals containing exponential factors in the E's and the magnitudes, especially of the E_D's, can be quite large, especially near the sinks so that a linear expansion of the exponentials can be quite inaccurate. Only for very small values of e_I° and e_V° would linear variation be expected.

4. Discussion

We have displayed various numerical evaluations of the swelling rates predicted by our model. Its chief virtue would appear to be that it has all physical features built into it which are felt to be important and thus eliminates the complex problems which have concerned various authors in their attempts to estimate and refine values for the various sink strengths and Z-factor which must be inserted into a reaction-rate-theory model. We have discussed several instances in which inconsistencies have also arisen in such analyses.

It is informative, however, to relate our void swelling rates here to the various "bias factors" employed in reaction-rate-theory analyses. To do this, let us return to Eq. (32) which we rewrite as

$$\delta \equiv \frac{d}{dt}\left(\frac{\Delta v}{v}\right) = I_{IC} - I_{VC} \tag{40}$$

where $I_C = 4\pi r_v^2 N_v J_c$ for both types of defects. Now conservation of vacancies

and interstitials independently, both with and without interaction fields, yields

$$-\mathring{a} = I_{IC} + I_{ID} = I_C(0) + I_D(0) \tag{41}$$

$$-\mathring{a} = I_{VC} + I_{VD} = I_C(0) + I_D(0) \tag{42}$$

since of course $I_C(0)$ and $I_D(0)$ are the same for vacancies and interstitials. Multiplying Eq. (40) by $\mathring{a}$ while dividing the two terms by $-(I_{ID} + I_{IC})$ and $-(I_{VD} + I_{VC})$, respectively (both of which are equal to $\mathring{a}$ by Eqs. (41) and (42)), we obtain

$$\delta = \left[\frac{I_{VC}}{I_{VD} + I_{VC}} - \frac{I_{IC}}{I_{ID} + I_{IC}} \right] \mathring{a} \tag{43}$$

which, after rearrangements, becomes

$$\delta = \frac{I_{VC}\, I_{ID} - I_{IC}\, I_{VD}}{(I_{VD} + I_{VC})\,(I_{ID} + I_{IC})} \cdot \mathring{a} \tag{44}$$

Now, if we introduce the Z-factors and use Eqs. (41) and (42) to convert the two factors in the denominators to currents in the absence of interaction fields, we obtain

$$\delta = \text{å}\,(Z_{VC}\,Z_{ID} - Z_{IC}\,Z_{VD})\,\frac{I_C(0)\,I_D(0)}{\left[I_C(0) + I_D(0)\right]^2} \qquad (45)$$

or

$$\delta = \text{å}\left[(Z_{ID} - Z_{VD}) - (Z_{IC} - Z_{VC})\right]\,\frac{I_C(0)\,I_D(0)}{\left[I_C(0) + I_D(0)\right]^2} \qquad (46)$$

with two forms of the "bias factor" which can be shown to be equivalent through Eqs. (3) and (4). The final factor in Eqs. (45) or (46) is completely determined by the sink structure and each I could equally well be replaced by the appropriate sink strength. Thus, Eq. (46) may be regarded as formally equivalent to Wiedersich's formula[6] except that he assumed his factors corresponding to $(Z_{ID} - Z_{VD})$ and $(Z_{IC} - Z_{VC})$ to be constants which they clearly are not. Nor are all of the Z's in Eq. (45) constant of course, as often assumed in reaction-rate-theory models. There is the possibility, however, that since the final factor in these very symmetrical formulas is a function of the sink strucutre alone, the bias factors containing the Z's may not depend strongly on sink structure. To simplify calculations of the bias factor, which we designate B, the factors $I_C(0) + I_D(0)$ may be replaced by å for our case of no recombination (although if one uses Eqs. (45) and (46) for estimates in which recombination is important this cannot be done). With this replacement then, Eq. (45) or (46) becomes

$$\delta = \text{å}BS_C(0)S_D(0) = \text{å}BS_C(0)\left[1-S_C(0)\right] = \text{å}BS_D(0)\left[1-S_D(0)\right] \qquad \text{(No recombination)} \qquad (47)$$

where $$S_C(0) \equiv -\frac{I_C(0)}{\dot{a}} = -\frac{4\pi r_v^2 N_v J_C(0)}{\dot{a}} \tag{48}$$

and $$S_D(0) \equiv -\frac{I_D(0)}{\dot{a}} = -\frac{2\pi r_o \rho\, J_D(0)}{\dot{a}} \tag{49}$$

and where $J_C(0)$ and $J_D(0)$ may be obtained from Eqs. (30) and (29) with $r = r_v$ and $r = r_o$ respectively, and with the thermal terms and all E's set equal to zero. We thus obtain

$$S_C(0) = -4\pi r_v^2 N_v \left\{ \frac{r_v}{3} - \frac{\left(\frac{2}{3}\frac{N_v}{\rho} R^3 + \frac{R^2}{2}\right) \ln\frac{R}{r_o}}{\left[r_v^2 \; 2\frac{N_v}{\rho} \ln\frac{R}{r_o} + \frac{1}{r_v} - \frac{1}{R}\right]} \right.$$

$$\left. + \frac{\frac{1}{4}\left(R^2 - r_o^2\right) - \frac{1}{6}\left(R^2 - r_v^2\right)}{r_v^2 \left[2\frac{N_v}{\rho} \ln\frac{R}{r_o} + \frac{1}{r_v} - \frac{1}{R}\right]} \right\} \tag{50}$$

With rearrangement, this can be written in the form

$$S_C(0) = \frac{4}{3}\pi N_v \left\{ -r_v^3 + R^3 \cdot \frac{\frac{2N_v r_v}{\rho} \ln\frac{R}{r_o} + \frac{3}{2}\frac{r_v}{R} \ln\frac{R}{r_o} - \frac{1}{4}\frac{r_v}{R} + \frac{3r_o^2 r_v}{4R^3} - \frac{1}{2}\frac{r_v}{R}^3}{\frac{2N_v r_v}{\rho} \ln\frac{R}{r_o} + 1 - \frac{r_v}{R}} \right\} \tag{51}$$

and, in similar fashion, we obtain

$$S_D(0) = \pi\rho \left\{ -r_o^2 + R^2 \cdot \frac{\frac{N_v r_v}{\rho}\left[\frac{4R}{3r_v} - 1 + \frac{2}{3}\left(\frac{r_v}{R}\right)^2 - \left(\frac{r_o}{R}\right)^2\right] + 1 - \frac{r_v}{R}}{\frac{2N_v r_v}{\rho} \ln\frac{R}{r_o} + 1 - \frac{r_v}{R}} \right\}$$

where the physical meaning of the S(0)'s as the fractional volumes from which the sinks drain defects is clearly displayed. One easy limiting case is $r_v \to 0$, for which $S_C(0) \to 0$, $S_D(0) \to \pi\rho(R^2 - r_o^2) + \frac{4\pi R^3}{3} N_v = 1$, the dislocations absorb all defects and of course no swelling occurs. The coefficients of R^3 in Eq. (51) and of R^2 in Eq. (52) determine the location of the surface around each type of sink across which the flux is zero in the absence of interactions and they are plainly related strongly to the relative sink strengths through the factor $N_v r_v/\rho$. The term $2N_v r_v \ln \frac{R}{r_o}/\rho$ is of course exactly the ratio of sink strengths obtained from simple, single-cell calculations. The $\frac{r_v}{R}$ and $\frac{r_o}{R}$ factors describe sink "volume-fraction" corrections and the presence of parameters of both types of sinks in each S(0) produces the so-called "multiple-sink" corrections discussed by other authors[8]. We feel that since these effects are contained within the single model in an internally consistent manner, they are probably more reliably described here than in other treatments where they are estimated through different models (always with approximations whose inaccuracies are difficult to assess) and then introduced into the reaction-rate-theory model as "correction factors". Unfortunately, we are still subject to the assumption that recombination can be adequately assessed separately, but all other models calculating sink strengths, bias factors or multiple-sink correction factors invoke the same assumption in addition to the lack of internal consistency discussed earlier.

Values of the bias factor, B, have been calculated from Eq. (47) for the various problems which we discussed above and are shown in Fig. 6. All unmarked

points are for T=600K, whereas numbers beside points designate other temperatures. The temperature dependence is very slight within the range of significant void swelling, but there is a systematic variation with relative sink strengths with B ~ 0.53 for equal sink strengths (maximum swelling). It falls to values of ~ 0.25 for the dislocation-dominated case and rises to ~ 1.5 for the void-dominated case.

These bias factors are much larger than the (Z-1) factors of 0.02 suggested by Brailsford and Bullough[7] or 0.08 deduced by Bullough, Eyre and Krishan[29] when matching their reaction-rate-theory predictions with experiment. However, these authors employed the full damage-rate predictions in their analyses and we believe this is incorrect. Although the actual fraction of defects which survive close-pair annihilation and thus are available to diffuse to sinks is difficult to assess quantitatively, it seems certain it can be significantly less than unity even for electron irradiation where close-pair annihilation typically removes[30] as many as ~80% of the defects under post-irradiation annealing in pure metals (but perhaps only ~10% in alloys). For neutron or heavy ion damage, cascade effects can reduce the number of defects surviving close-pair annihilation even further. We recently concluded[20] that fewer than ~18% of the defects escape a typical neutron cascade; Blewitt et al[31] and Goldstone et al[32] have interpreted independent experimental measurements to mean that only ~1% of the point defects escape a high-energy cascade. It seems clear that the effective production rates for neutron damage can easily be only ~0.1 of the TRN standard calculations. Then if previous authors estimated a required bias of ~0.06 using the full damage rate, a more

reasonable estimate would be $B \gtrsim 0.6$ which is very close to the average of our estimates. An alternative way of expressing the situation is that our estimates are consistent with experimental observations and our knowledge of close-pair annihilation whereas the bias values of a few percent are not.

Fisher and White[33,34] have previously suggested that the larger bias factors predicted by the Heald single-cell approach offer a better explanation for observed swelling rates (when the effective defect production rate is considered as we have done here) than does the small bias factor proposed by Bullough and co-workers. We concur with this general conclusion, but we consider that our present bias estimates should be more quantitatively reliable than those derived from Heald's model which considers only dislocations to derive Z_{ID} and Z_{VD}, inconsistently assumes $Z_{IC} = Z_{VC} \equiv 1$ and simulates defect production by imaginary sources outside the dislocation cell boundary. For example, we find B increases from ~0.29 to ~0.46 as N_v goes from ~10^{19} to ~10^{21} m^{-3} ($r_v = 100\ r_o$) for a <u>fixed</u> dislocation density of 6×10^{14} m/m^3. The Heald model of course has no effect of void sink strength. Also, we find that for a fixed void density of 10^{21} m^{-3} ($r_v = 100 r_o$) B decreases from ~1.55 to ~0.46 as the dislocation density increases from 10^{10} to 6×10^{14} m/m^3. Heald's approach yields values of B which <u>increase</u> over the same range from ~0.15 to ~1.15. Thus not only do the magnitudes differ but they vary in the <u>opposite</u> direction as a function of dislocation density.

Finally, we compare our results with those predicted by Wolfer and Ashkin[13,16]. These authors claim that cavities atract interstitials preferentially more than do dislocations so that void growth cannot even occur unless the cavities are first "coated" with segregated solute atoms which set up additional interaction fields which <u>repel</u> the interstitial more than the vacancy. Now certainly solute segregation to voids (and other sinks) does occur and this phenomenon can produce additional interaction fields which in turn will alter the bias. Indeed, such

effects could easily be incorporated into the present model by specifying these interaction fields in addition to the intrinisic ones used here. However, we reject their contention that "clean" voids will not grow in the presence of "clean" dislocations. They arrived at this conclusion by eliminating the first-order misfit interaction between a dislocation and a point defect on the basis that its average over the full angular range is identically zero. This is clearly inconsistent with the fact that, in analytical studies in the absence of irradiation, the fully angular dependent interaction has been shown[25] to be equivalent to an interaction independent of angle with a magnitude equal to the actual interaction averaged over only the attractive regime (exactly our assumption). The prediction by Wolfer and Ashkin[16] of a much stronger effect of externally applied tensile stresses than that predicted here also arises from the neglect of the misfit interaction contribution to the interaction energy, so we believe them to be in error on this point also.

5. Summary

We have developed an analytical approach which considers the production of point defects by irradiation and their migration to different types of sinks with which they interact in differing ways. We have used this approach to analyze the resulting bias effects, or the absorption of relatively different amounts of one type of defect at particular sinks. This bias effect results in radiation-induced dislocation climb which figures prominently in current concepts of radiation-induced creep, growth and swelling. However, all prior models to estimate the magnitudes of these bias effects suffer from definite errors or unphysical assumptions which render them of uncertain quantitative accuracy.

The approach presented here combines together, for the first time, defect production and partitioning among more than one type of sink with different interaction fields. We have applied it to obtain specific quantitative estimates of the magnitude of SIPA creep and found its rate to be significantly less than previous estimates, the magnitude of the bias factor being ~0.07% for an applied stress of 10^2 MPa and dislocation density of 6×10^{14} m/m^3. This value is weakly temperature-dependent and increases with increasing dislocation density.

We have also applied it to the case of dislocations and spherical voids of arbitrary densities. The predicted bias factor for assumed values of 1.4 and 0.23 for the relaxation strains of interstitials and vacancies, respectively, is ~0.6 for comparable magnitudes of the dislocation and void sink strengths, falls to ~0.26 for the dislocation-dominated case and rises to ~1.5 for the void-dominated case. These magnitudes have been shown to be consistent with experimental results and our present understanding of the number of point defects surviving close-pair annihilation following their production. Previous theoretical estimates of bias factors of only a few percent (or in one case even negative values) have been shown to be theoretically deficient, and previous estimates of a few percent deduced from experimental results employed the unreasonable assumption that <u>all</u> the defects calculated to be produced by a model such as the TRN standard are available for long-range diffusion.

Estimates have also been made of the effect of externally applied stress and gases inside the voids on the resulting swelling and it was concluded that pressures ~10^{-3} of the shear modulus would be required for significant effects.

The possibility of applying such high pressures without yielding is problematical so that observation of stress-enhanced swelling effects seems difficult if not unlikely.

The present model enables the calculation of more internally consistent and therefore probably more accurate volume-fraction and multiple-sink correction factors for sink strengths than those previously available.

Our model has ignored recombination, thus implying that an effective defect production rate can be employed which has been corrected for losses due to recombination. The latter must, in turn, be independently estimated and so we have no guarantee that errors in predicted mass-transport rates are not thereby introduced. However, the recent analysis of Hayns[35] indicates that these errors are likely to be quite minor in most cases.

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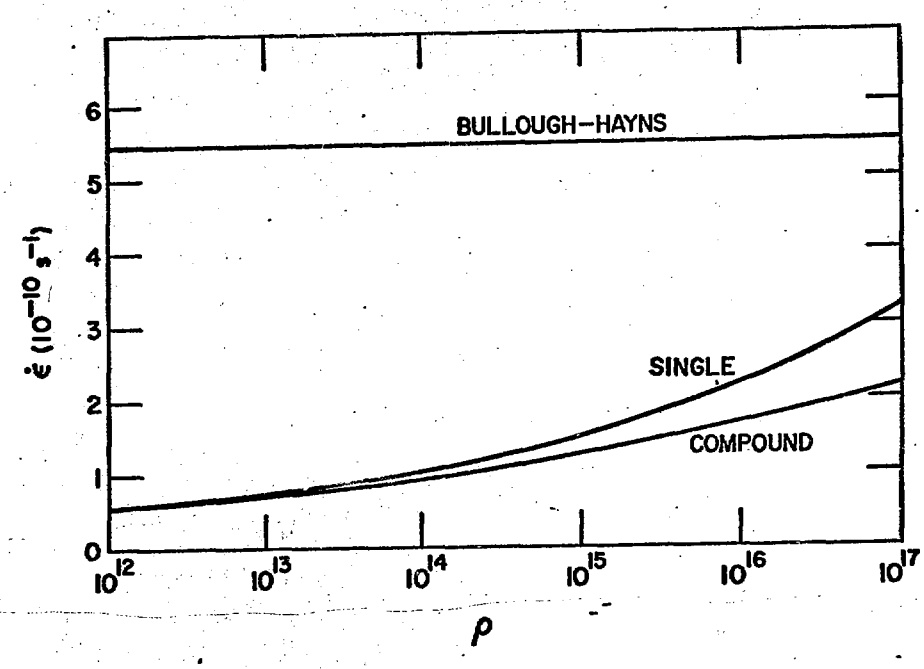
List of Illustrations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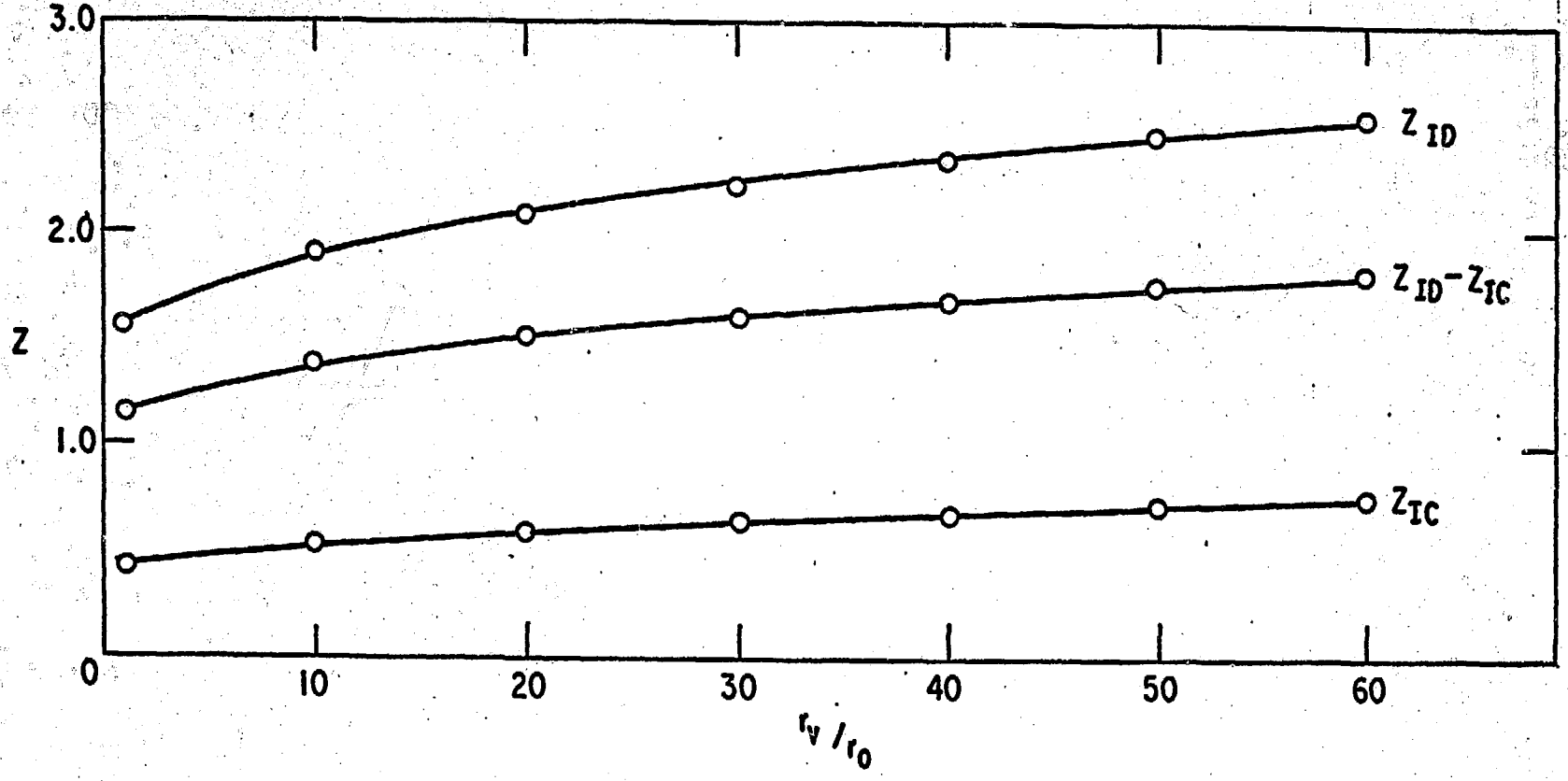
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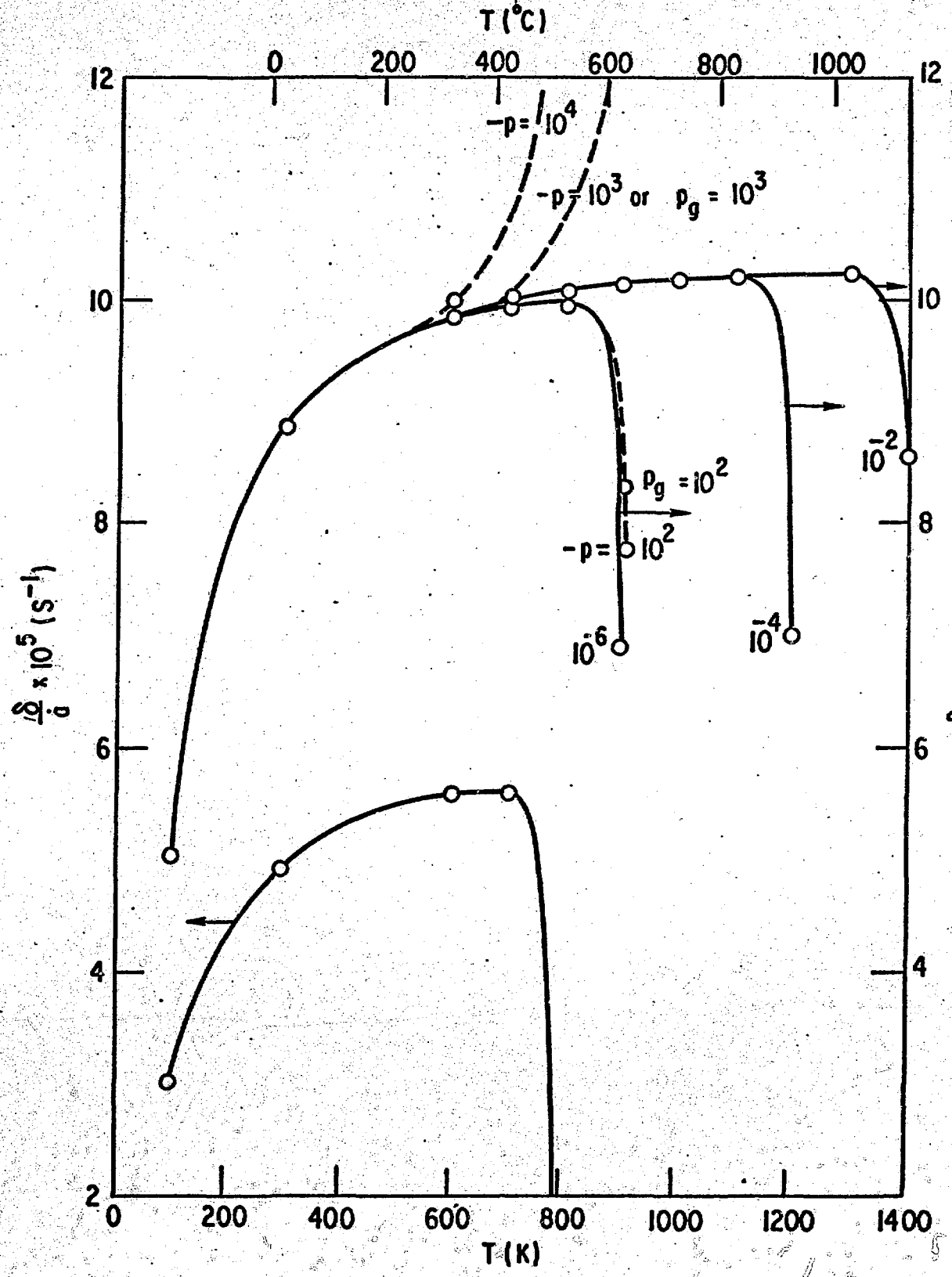
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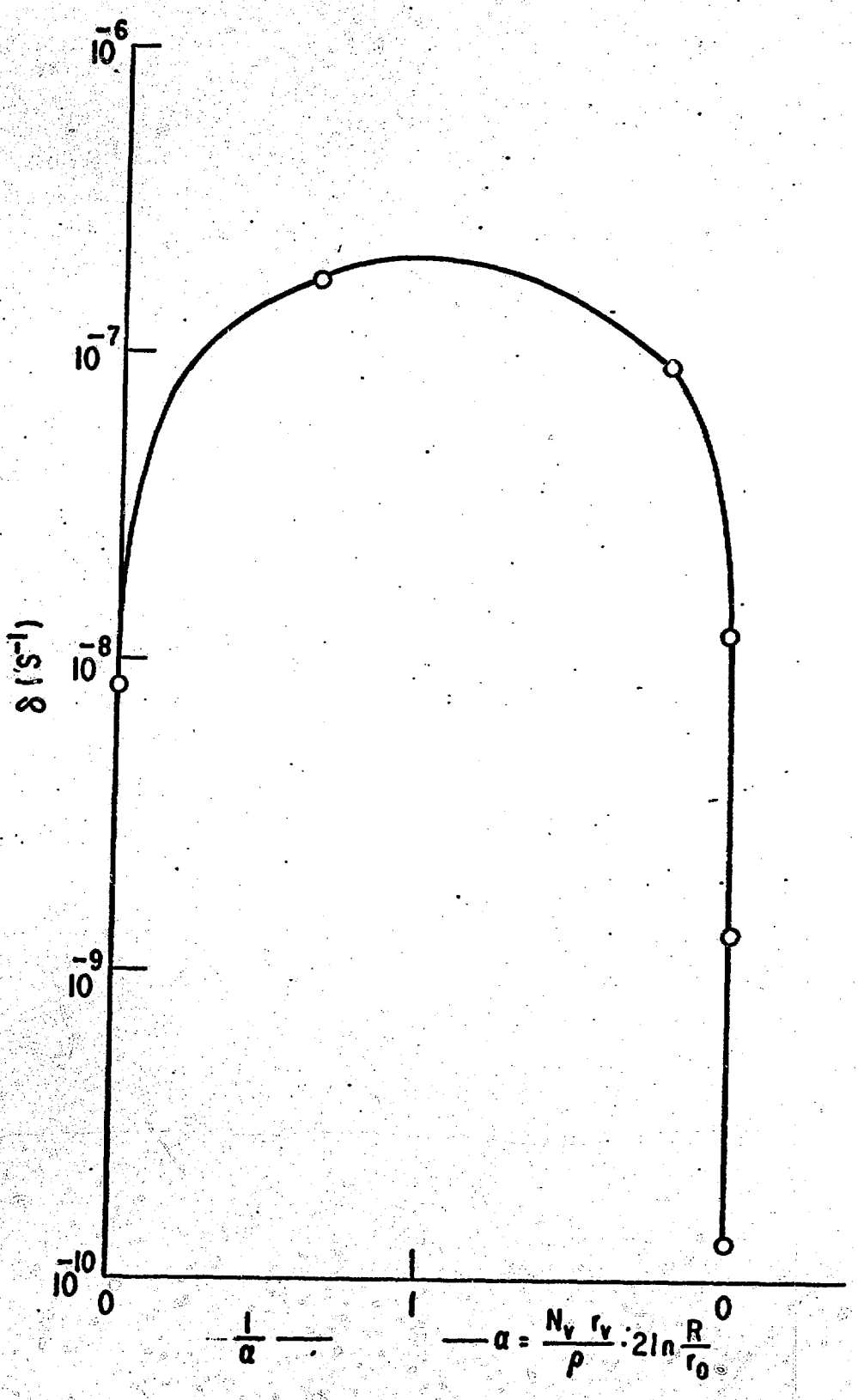
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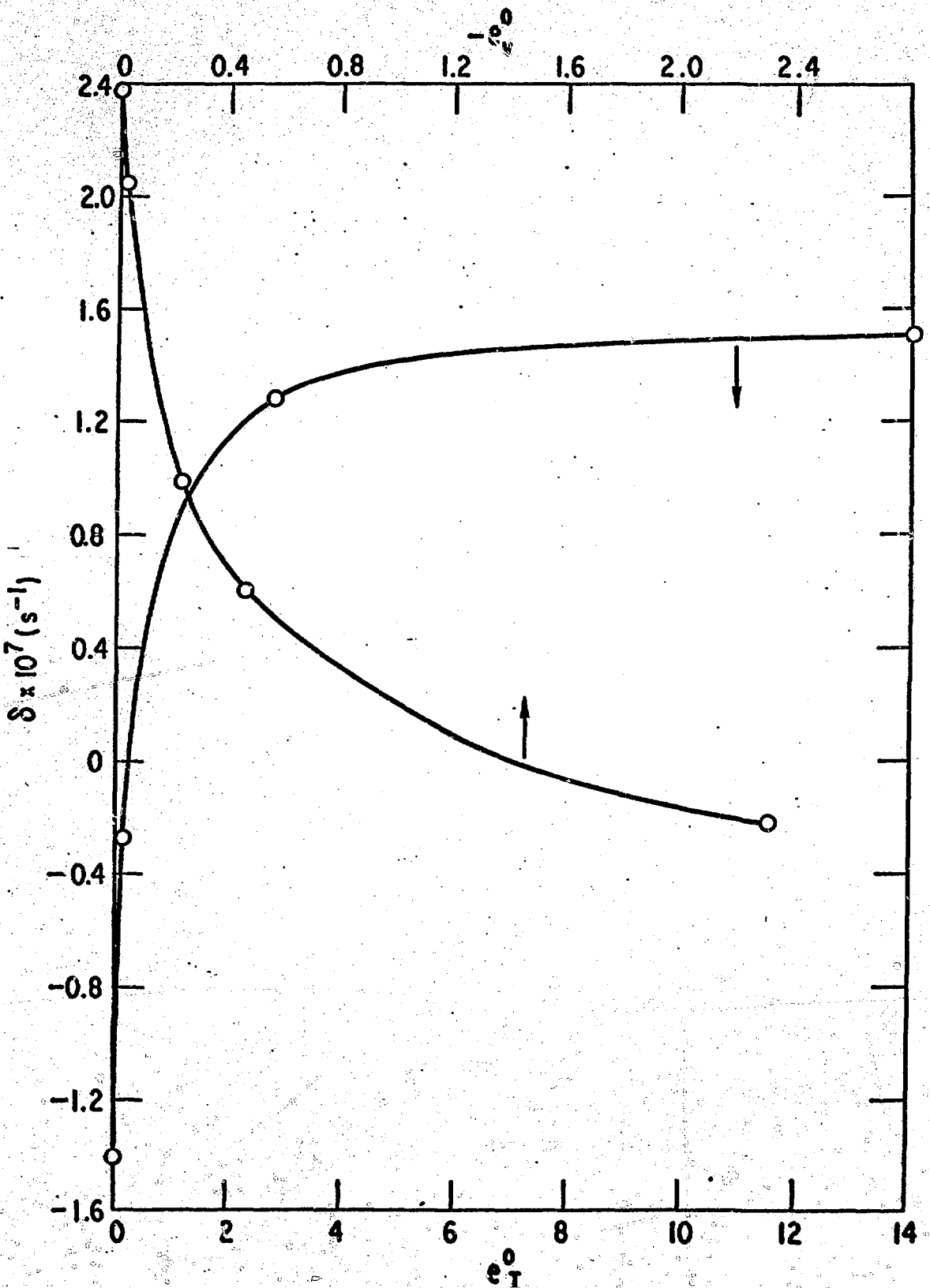
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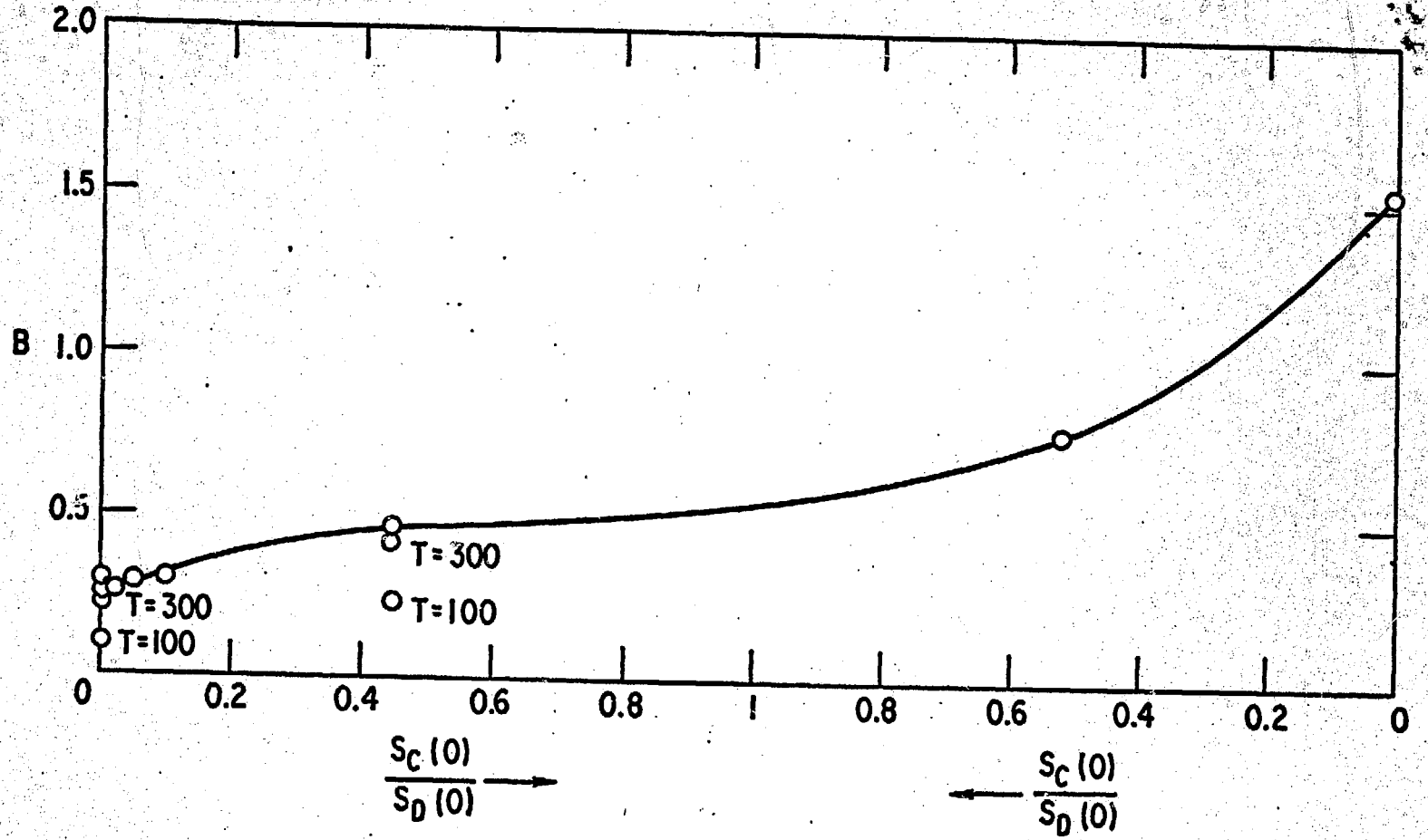
14
e^0_T


2.0
1.5
B 1.0
0.5
0
T=300
T=100
T=300
T=100
0.2
0.4
0.6
0.8
1
0.8
0.6
0.4
0.2
0
$\frac{S_C(0)}{S_D(0)}$ →
← $\frac{S_C(0)}{S_D(0)}$