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STRESS-INDUCED DEFORMATION OF METALS DURING FAST-NEUTRON IRRADIATION*

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ABSTRACT

Theoretical models for the deformation of metals subjected simultaneously to external loads and fast-neutron irradiation are discussed. The following models have been analyzed in detail: dislocation climb; climb controlled glide; nucleation, growth and unfaulting of dislocation loops. It is shown that the strains produced only by the climb of dislocations are equal to the isotropic swelling strains plus the thermal creep strains due to climb of dislocations. No irradiation-induced creep results from the climb of dislocations or growth of loops.

The climb controlled glide of dislocations is a viable irradiation creep mechanism for low fluences at all temperatures or for all fluences at high temperatures. At low temperatures the hardening occurs by the elastic interaction of loops and dislocations. It is shown that the climb controlled glide of dislocations through the loop structure can explain the transient irradiation creep behavior.

Steady-state creep, however, at low temperatures is explained by the stress-affected nucleation of dislocation loops. Comparison of the theoretical prediction with the experimental results suggests that stress not only influences the orientation of the loop nuclei but also enhances the interstitial loop nucleation and reduces the vacancy loop nucleation. In addition, various mechanisms are discussed briefly for the effects of stress on the irradiation-induced swelling of metals. Finally, since irradiation creep and swelling both occur by partial segregation of vacancies and interstitials it was concluded that the two phenomena are closely related.

I. INTRODUCTION

Metals which are subjected simultaneously to external loads and fast neutron irradiation may experience two particular modes of deformation: irradiation-induced creep and stress-affected swelling. Of the two only the former has been measured experimentally for various materials. Stress-affected swelling, however, has been postulated based on theoretical models as well as indirect evidence from irradiated fuel pins.[1,2,3,4]

In the past, various models have been proposed for irradiation-induced creep.[5] Those applicable to fast neutron irradiation and cubic metals can be divided into two categories. In the first group, irradiation-enhanced climb is considered to be responsible for creep, although much controversy has arisen about this mechanism.[6,7,8] In the second category, irradiation creep is associated with the formation and growth of point defect clusters.

In the present paper we briefly review the mechanisms for irradiation creep and stress-induced swelling and report on some conclusions from recent theoretical work concerning these subjects.[2] The discussion will include dislocation climb, climb controlled glide of dislocations, stress-induced loop alignment, and stress-assisted swelling.

In a recent paper,[2] it was proposed that a relationship exists between swelling and irradiation creep. Additional theoretical aspects of this relationship will be explored in this paper. An experimental method for defining this relationship involving slit tube tests was proposed by Pennell,[9] and an analytical examination of this method was presented in another paper in this conference[10]. One practical result to be expected from a direct relationship between the irradiation creep rate and the swelling rate, is that differential swelling would not be capable of generating large stresses in core components.

II DISLOCATION CLIMB

In the first attempts to explain irradiation creep it was assumed that an enhanced climb of dislocations under irradiation would necessarily lead to creep deformation.[11] This view is shared by many researchers, although Mosedale,[12] Hesketh[13] and Wolfer et al.[2] have shown that the stress-induced differential climb of edge dislocations is not affected by irradiation. This is illustrated in Figure 1. In the absence of stress all edge dislocations would climb the same distance α per unit time if the metal swells. If a uniaxial stress is now applied, edge dislocations or loops whose Burgers vector b is parallel to the direction of the applied stress, climb an additional distance β per unit time. Edge dislocations with Burgers vectors perpendicular to the stress do not change their climb rate.

An alternative way to describe the stress-enhancement of climb is shown on the right hand side of Figure 1. All edge dislocations may be considered to have climbed the distance $\alpha + \frac{1}{2}\beta$ first which means that the swelling has increased by the application of the stress. The additional term can then be shown to be identical to the thermal creep due to the climb of dislocations which is independent of the irradiation. The increase in the climb rate under stress is due to the fact that the vacancy concentration at properly oriented dislocations is enhanced. The reduced flow of vacancies to the dislocations is of course accompanied by an increased flow of vacancies to the voids. The enhancement of swelling due to this process is closely connected with thermal creep due to dislocation climb.

In fact, it can be shown that the total thermal creep rate must obey the relation[14)]

$$\frac{\dot{\varepsilon}_1}{\sigma_1-\sigma_H} \gtrsim \frac{2\pi}{15\ln(R/r_v)} \frac{\rho}{nr_o} \frac{(\frac{\dot{\Delta V}}{V})_\sigma - (\frac{\dot{\Delta V}}{V})_o}{\sigma_H}, \tag{1}$$

provided the enhancement of the swelling rate by the stress, $(\frac{\dot{\Delta V}}{V})_\sigma - (\frac{\dot{\Delta V}}{V})_o$ is small compared to the stress-free swelling rate, $(\frac{\dot{\Delta V}}{V})_o$. In the above equation, ρ is the dislocation density, 2R their average spacing, r_v the capture radius for vacancies, n the void density, and r_o the void radius obtained in a stress-free metal. The equation (1) may be used in the interpretation of the observed diameter changes in fuel pins. If it is observed that $\Delta D/D$ is identical to $1/3\ \Delta V/V$, there can be no stress-enhancement of swelling according to the above mechanism.[14,15)] It is, however, conceivable that other stress-assisted processes enhance swelling indirectly. This topic is discussed further in Section V.

III CLIMB CONTROLLED GLIDE OF DISLOCATIONS

Since the climb of dislocations by itself can not account for irradiation creep, other possibilities for irradiation creep must be considered. One obvious choice is the climb controlled glide of dislocations, and various models of irradiation creep based on this idea have been developed in the past.[16,17,18)] The basic process of all these models is the irradiation-induced glide of dislocations over obstacles in their glide planes. However, two basic difficulties are encountered.

First the glide can be so severely restricted by the irradiation produced loops that no creep may take place. For example, if ℓ is the average spacing of the loops, and R their radius, the stress must be larger than

$$\sigma \geq 4Gb \frac{R^2}{\ell^3} , \qquad (2)$$

where G is the shear modulus and b the Burgers vector. Since $\ell = 0.55\ N_\ell^{-1/3}$, where N_ℓ is the loop density,

$$\sigma \geq 8G \left(\frac{\Delta V}{V}\right)_{loops} , \qquad (3)$$

for $\pi R^2 b\ N_\ell$ is the volume fraction $(\Delta V/V)_{loops}$ occupied by the atoms contained in the loops. Stiegler and Bloom have observed[19)] that after a fluence of about 10^{22} nvt the interstitial loops in solution-annealed type 304L stainless steel occupy a fractional volume of about 0.1%. Thus, the minimum stress required for dislocations to glide past the loops is approximately 80,000 psi. For this reason, we believe that climb controlled glide will not contribute to irradiation creep at fluences above 10^{22} nvt and at low temperatures where a high loop density is formed. However, the mechanism may occur at higher temperatures, where a large number of loops can not be sustained, or it may occur at all temperatures for very low fluences.

The second difficulty referred to above involves the dislocation climb rate. In the absence of sinks with different bias, i.e. different interactions with the point defects, the dislocations cannot climb. Should, however, voids be present, then the climb rate of the dislocations is simply proportional to the swelling rate of the metal.[2,18] The frequency with which dislocations escape the glide obstacles is then equal to the climb rate divided by the average obstacle height d. If L is the average distance between the glide obstacles the creep rate is then given by[2)]

$$\dot{\varepsilon}_1 = \frac{\sqrt{3}}{2}\frac{L}{d}\;\frac{\sigma_1-\sigma_H}{\sigma_{eq}}\;(\frac{\dot{\Delta V}}{V}), \tag{4}$$

where $\sigma_H = \frac{1}{3}\;(\sigma_1 + \sigma_2 + \sigma_3),$ (5)

and $\sigma_{eq} = \frac{1}{\sqrt{2}}[(\sigma_1-\sigma_2)^2 + (\sigma_2-\sigma_3)^2 + (\sigma_3-\sigma_1)^2].$ (6)

Equation (4) predicts that the creep due to climb controlled glide of dislocations is proportional to the swelling rate. This result was derived under the assumption that all dislocations exhibit the same bias for interstitials and thus climb at the same rate. If no swelling occurs, the irradiation creep must cease according to equation (4).

Nevertheless, it is in principle possible, to obtain irradiation creep by climb controlled glide even in the absence of swelling. For this to occur some dislocations must climb in one direction, and other dislocations in the opposite direction. The required partial segregation of vacancies and interstitials is indeed possible because of the following reason. It has been shown[20)] that isolated dislocations and dislocations arranged in dipole configurations exert a different bias on the point defects. The reason for this difference is simply that the stress field of an isolated dislocation has a $1/r$ dependence, whereas the stress field of a closely spaced dipole decreases as $1/r^2$ with the distance r. Therefore, in an inhomogeneous dislocation network, one expects interstitials to flow preferrentially to isolated dislocations whereas dislocation tangles absorb more vacancies and thereby cause a differential dislocation climb. In such a case, the factor $(\frac{\Delta V}{V})$ in equation (4) is replaced by the climb rate of either group of dislocations.

We expect that this differential dislocation climb is more pronounced in cold-worked than in annealed materials, since cold-working produces a highly non-uniform dislocation structure. Furthermore, differential dislocation climb is expected to decrease with fluence as a more uniform dislocation structure is regenerated by the unfaulting of dislocation loops which seem to be continuously produced by the irradiation. Therefore, we conclude that climb controlled glide of dislocations occurs mainly during the transient period of irradiation creep. Because of the argument given above, transient creep should be more pronounced in cold-worked materials. This is indeed observed, as discussed in more detail by Foster et al.[10]

In order to find the stress-dependence of irradiation creep by climb controlled glide it is important to identify the major glide obstacles. It has been shown by Makin et al.[21] that irradiation hardening in metals is caused by the dislocation loops. Therefore, we conclude that loops also represent the dominant glide obstacles for irradiation creep. Based on this premise, Wolfer et al.[2] have estimated the stress dependence of the creep rate given by equation (4) and obtained a relation $\dot{\varepsilon}_I \sim \sigma^{1.2}$. However, a more refined analysis is given below.

Because the stress field at a distance r from a loop of radius R is[22)]

$$\sigma \cong Gb \frac{R^2}{2r^3} , \tag{7}$$

a dislocation cannot bypass a loop if the distance of closest approach is smaller than

$$r = [\frac{GbR^2}{2\sigma}]^{1/3} , \tag{8}$$

where σ is the external stress. The average separation of stress-fields intersecting the glide planes is given by[23)]

$$L = \frac{1}{2} (2rN_{\ell})^{-1/2} . \tag{9}$$

To escape the influence of the stress-field of a loop the dislocation has to climb an average distance

$$d = r/2. \tag{10}$$

Therefore,

$$\frac{L}{d} = (2r^3N_{\ell})^{-1/2}, \tag{11}$$

and with equation (8) and the relation $(\frac{\Delta V}{V})_{loops} = \pi R^2 N_{\ell} b$ we obtain

$$\frac{L}{d} = \{ \frac{\pi\sigma}{G(\Delta V/V)_{loops}} \}^{1/2} \tag{12}$$

This result is only valid for low stresses. At high stresses, dislocations will tend to pile up and the stress exerted by the front dislocation is then equal to $(2\ell_p \sigma^2/Gb)$, so that L/d becomes

$$\frac{L}{d} = \left\{\frac{2\pi\ell_p}{b(\Delta V/V)_{loops}}\right\}^{1/2} \frac{\sigma}{G} \tag{13}$$

where ℓ_p is the length of the pile-up.

Combining equations (4), (12), and (13), irradiation creep by climb controlled glide, or transient creep, is thus expected to be proportional to $\sigma^{1/2}$ at low stresses but to become linearly dependent on the stress for higher stresses. Again, this is in qualitative agreement with the experimental observations on the stress dependence of transient irradiation creep.[10,24] Since transient creep is either terminated because of loop hardening or because the loops eventually create a more uniform dislocation structure, the transient period is expected to be determined by the time period t_m which is necessary to grow loops to their maximum radius R_m. Under typical neutron conditions the loop growth rate is of the order of 5×10^{-12} cm/sec.* If we assume that $R_m \cong 300$ Å, the time $t_m \cong 170$ hours. Thus, we expect the transient period to extend to fluences of about 10^{21} n/cm^2, which is in agreement with experimental observations.[24,26]

* This can be estimated with the formula $dR/dt \cong b^2(Z_i - Z_v)\sqrt{P/\alpha}$, where P is the production rate of Frenkel pairs, $\alpha = 504b/D_v$ with b equal to the Burgers vector, and D_v equal to the vacancy diffusion coefficient. The difference in the bias factors Z_i and Z_v is approximately equal to 0.01.[25]

IV NUCLEATION AND GROWTH OF DISLOCATION LOOPS

Irradiation of stainless steels in the high voltage microscope has revealed that loops are nucleated continusouly below 500°C at least to a dose of 6 dpa.[27] Dislocation loops are also found in neutron irradiated materials in approximately the same temperature range together with voids. Fisher and Williams[28] have recently explained the dose dependency of swelling observed under ion, electron, and fast neutron bombardment. They postulated that there are three distinct stages pertaining to the evolution of the loop structure. In the first stage loops nucleate and grow continuously and the dislocation density builds up. In the second stage a dynamic equilibrium is reached between nucleation and growth of loops and their mutual annihilation. Finally, in the third stage, nucleation of loops ceases and the dislocation density decreases at high doses. Under neutron irradiation the third stage can probably not be observed because of the relatively low dose rate.

In metals exhibiting extensive void growth, mainly interstitial loops are observed. However, both types of loops have been identified in metals irradiated at temperatures where swelling is possible. It appears that the vacancy loops are more numerous but smaller than the interstitial loops. Once stable three-dimensional vacancy clusters are produced through gas-stabilization, vacancy loop formation apparently ceases.

According to recent theories developed by Russels,[29] Burton,[30] and Katz and Wiedersich[31], the nucleation of voids and loops is mainly governed by kinetic rather than by thermodynamic factors. These theories suggest that as far as the effect of stress is concerned the nucleation rate is given by

$$\nu = \nu_o \exp \{\pm \pi R_o^2 b \sigma_n / kT\}, \qquad (14)$$

where ν_o is the stress-free nucleation rate, R_o the effective radius of the critical loop and σ_n the stress component normal to the loop area. The plus sign is valid for interstitial loops and the minus sign for vacancy loops. Equation (14) states that stress influences the orientation of the nucleating loops as well as their number. Although there is little doubt about the orientation dependence, stress may have no effect of the total nucleation rate. To illustrate this point we have calculated the creep rate in a torsion experiment as a function of the shear stress for the two limiting cases where stress does or does not enhance the total nucleation rate of loops. Without giving the detailed calculations, the results are as shown in Figure 2. In the latter case the creep rate will eventually become independent of the stress when all loops are positioned in their energetically most favorable orientation. In both cases, however, the creep rate is proportional to τ for low values of the shear stress. Actually, the extent of the linear range depends on the value of $\pi R_o^2 b\tau/kT$, and thus on the critical radius R_o. Since $\pi R_o^2 b$ is the effective volume of the critical loop we may also write $\pi R_o^2 b = n\Omega$, where n is the effective number of defects in the critical loop and Ω the atomic volume.

If the critical loop size were just one interstitial, n can be estimated from the difference of the dipole tensor component of the dumbell interstitial, as calculated by Seeger et al.[32] In this case one finds that n(interstitial) $\cong$ 1/2. As the critical size increases n rapidly approaches the actual number of interstitials in the cluster. Since interstitials strongly interact and bind each other, and since the migration energy of these clusters rapidly increases with the size[33], we estimate that the critical cluster size consists of three to six interstitials. Thus, we expect that $n\Omega\sigma_n/kT$ becomes of the order of one and that a deviation from linearity occurs for stresses between 10,000 and 20,000 psi. This is indeed observed in the irradiation creep of 20% cold-worked M316 at equivalent stresses above 15,000 psi.[10,24] Furthermore, since the deviation from linearity is such as indicated by the curve A in Figure 2, we believe that stress also influences the total nucleation rate of loops, i.e. tensile stresses increase the nucleation rate of interstitial loops and reduce the nucleation of vacancy loops. This behavior may then be interpreted as a stress-enhancement of swelling.

To show the magnitude of the creep rates due to loop nucleation the loop volume ($\pi R_m^2 b\nu_o$) produced per unit time has to be evaluated. Brager and Straalsund[34] have recently reported detailed measurements on loop density and size for solution-annealed type 316 stainless steel. At an irradiation temperature of 390°C and a fast fluence of 0.75×10^{22} (E>0.1 Mev), the observed loop density was 10^{16} cm^{-3} with an average diameter of 250Å. According to Figure 2 the linear portion of the irradiation creep rate is given by:

$$\dot{\gamma} = \frac{2}{3} \left[\frac{\pi R^2 b\nu_o t}{\phi t}\right] \left[\frac{\pi R_o^2 b}{kT}\right] \phi\tau. \qquad (15)$$

Equation (15) can be rewritten as:

$$\dot{\gamma} = \frac{2}{3} \left[\frac{(\frac{\Delta V}{V})_{loop}}{\phi t}\right]\left[\frac{n\Omega}{kT}\right] \phi\tau, \qquad (16)$$

and evaluated on the basis of Brager and Straalsunds[34] data as follows:

$$\dot{\gamma} = \frac{2}{3} \left[\frac{1.15 \times 10^{-3}}{0.75 \times 10^{22}}\right] 1.1 \times 10^{-5} \; n\bar{E}\phi\tau \cong 1.1 \times 10^{-30} \; n\bar{E}\phi\tau. \qquad (17)$$

This constant is in good agreement with the experimental value for solution-annealed M316 of 4.7×10^{-30} obtained by Foster, et. al.[10] from an analysis of the data reported by Mosedale and Lewthwaite.[24]

It should be noted that irradiation creep due to stress-affected loop nucleation requires only the existence of one type of loop. Should vacancy loop formation not occur, one would expect that the volume of the interstitial loops is at least a fraction of the irradiation-induced volume change. It has in fact been demonstrated by Stiegler and Bloom[19] that the observed density change in type 304L stainless steel can be accounted for by the interstitial loop volume. Since irradiation creep due to stress-affected loop nucleation is also proportional to the loop volume, irradiation creep and swelling are interrelated. Irradiation creep by this mechanism does not necessarily require the existence of swelling if interstitial and vacancy loops are formed simultaneously. Other possibilities which do not involve volume changes require that one type of defect forms loops, and the other type is absorbed at dislocations.

The existence of both types of loops has been observed in ion-bombarded type stainless steel by Laidler,[35] and in molybdenum by Brimhall et al.[36] However, when swelling occurred in these materials the vacancy loops could no longer be observed. This result verifies the contention that vacancy loops form by the collapse of three-dimensional clusters, and that when the collapse is prevented, swelling takes place.

Microstructural observations of irradiated metals consistently reveal that loops can not be observed above a critical irradiation temperature which is of the order of $0.5T_m$. The critical temperature appears to be lowered by cold working. If indeed no loop formation occurs above this critical temperature, irradiation creep due to loop alignment will cease to exist. In such a case, however, irradiation creep probably proceeds by the climb controlled glide mechanism discussed in the last section.

It has been suggested that external stresses may influence the direction of the unfaulting of Frank loops and thus cause a contribution to irradiation creep.[37] As discussed by Wolfer et al.[2] this contribution should become independent of stress above relatively small stress values. In addition, Bullough et al.[38,39] have shown that a rotation of the loops occurs after unfaulting. The rotation causes the loop normal to become again parallel to the Burgers vector and thus cancel the strains produced by the unfaulting. Therefore, we do not expect that the unfaulting of loops makes any contribution to irradiation creep.

V. THE EFFECTS OF STRESS ON SWELLING

The effects of stress on swelling were discussed earlier in connection with dislocation climb. In this section, other possible effects of stress will be examined. In the past, two different effects have been considered in some detail that could give rise to an enhancement of swelling under tensile stress. At temperatures above $0.5T_m$, an exchange of vacancies between the voids and the dislocations takes place that leads to volumetric changes as well as volume-conservative changes.[1,2,3] Straalsund et al.[15] have expressed this effect in terms of an effective Poisson's ratio which deviates from 1/2, the value consistent with volume-conservative creep. A different, but equivalent description of this effect was described earlier in Section II.

The stress-induced increase of the void volume by exchange of vacancies does not depend directly on the irradiation, and should more correctly be described as a stress-effect on the annealing rate of voids.[2,3,15] Figure 3 shows the enhancement of swelling due to this mechanism for a tensile stress of 20,000 psi. It is seen that this effect is only important at high temperatures.

At low temperatures, a possible effect of stress on void growth is the change of the vacancy migration energy. However, it has been shown[2] that this effect would lead only to a small change of the void growth rate, as shown in Figure 3.

If the external loads produce deviatoric stress components, the external stresses incude an elastic interaction between the interstitials and the void due to the non-uniform stress distribution around the void. Li et al.[40] and Bullough and Perrin[41] have estimated this effect approximately and apparently arrived at opposite conclusions. According to Li et al. uniaxial tensile stresses reduce the void growth rate whereas Bullough and Perrin find that compressive stresses reduce it.

A complete analysis of this effect should be expressed in terms of deformation strains since stress-affected swelling is most probably connected with creep strains. It is suspected that the different results obtained by Li et al. and by Bullough and Perrin can be reconciled if this is done.

Other possible effects of stress on swelling are:

a) stress-induced loop nucleation,
b) stress-assisted precipitation of carbides, and
c) stress-induced rearrangement of the dislocation network.

The influence of stress on interstitial and vacancy loop nucleation was discussed in the previous section. An enhancement of the former and reduction of the latter by tensile stresses is suggested by the irradiation creep results. Such an enhancement of irradiation creep would be accompanied by a corresponding enhancement of the swelling.

According to the observations of Appleby[42] enhanced swelling measured in irradiated fuel pin cladding is closely associated with the precipitation of carbides. He suggested that, stress may indirectly enhance swelling by accelerating the precipitation processes.

A very effective way by which stress could enhance the swelling is the change of the dislocation bias.[20] Under external loads it is expected that dislocations form pile-ups. A pile-up of edge dislocations can be considered as a single dislocation of strength Nb, where b is the Burgers vector and N the number of dislocations in the pile-up. Such a pile-up has then a capture radius which is N times larger than a capture radius of a single dislocation. Therefore, this type of dislocation configuration should provide a stronger preferential attraction for interstitials, and thereby increase the driving force for swelling.

VI CONCLUSIONS

1) Irradiation creep can proceed by either the climb controlled glide of dislocations or by the alignment of loop nuclei.

2) Irradiation creep by the climb controlled glide of dislocations can be responsible for the transient irradiation creep. Since it is driven by the difference in the bias of a non-uniform dislocation network, cold-worked materials show larger transient creep strains than annealed materials. The stress-dependence of the transient creep is $\sigma^{1/2}$ at low stresses and σ at high stresses. This mode of creep is terminated by a dense loop structure and by a uniform dislocation network.

3) At temperatures above $0.5T_m$ where a high dislocation density can not be maintained, the irradiation creep rate due to climb controlled glide is proportional to the swelling rate.

4) In the temperature range where loop nucleation occurs, the steady-state component of irradiation creep is consistent with the loop alignment model. The stress-dependence is linear at lower stresses but shows a more rapid increase at high stresses. This deviation from linearity indicates that tensile stress enhance the interstitial loop nucleation and reduces the vacancy loop nucleation. Electron microscopy observations indicate that when swelling occurs only interstitial loops are formed, and that the loop volume is of the order of the void volume. Thus, irradiation creep by interstitial loop alignment is expected to be related to swelling.

5) Possible effects of stress on swelling may be due to a number of processes. At high temperatures ($T>0.5T_m$) an exchange of vacancies between voids and dislocation is induced by hydrostatic tensile stresses. Furthermore, carbide precipitation may be accelerated by tensile stresses. Stress-effects which may alter swelling at lower temperatures are the stress-induced elastic interaction of the point defects with the voids and the formation of dislocation pile-ups.

6) Theoretical studies of irradiation processes indicate that irradiation creep and swelling are both produced by the partial segregation of vacancies and interstitials. This leads to the conclusion that the irradiation creep rate should increase as the swelling rate increases. One practical result of this relationship would be that differential swelling would not be capable of generating large stresses in core components.

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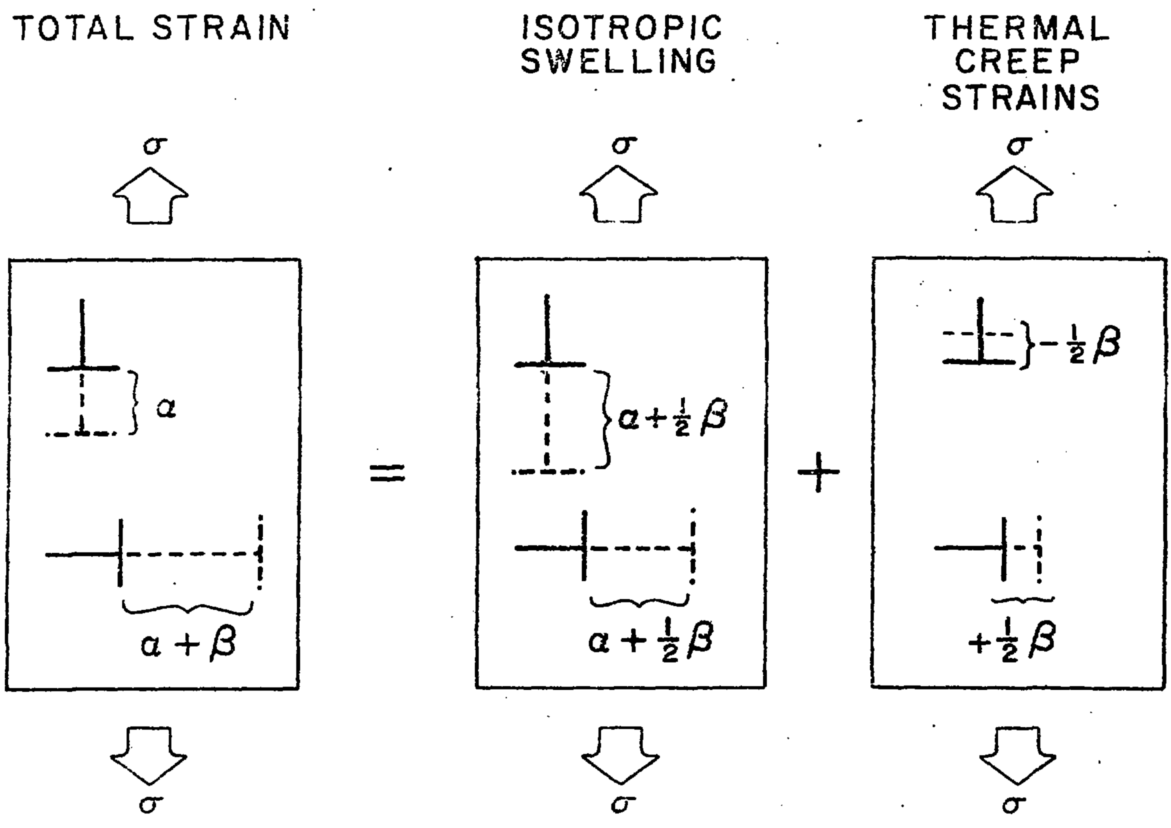
Figure 1

DISLOCATION CLIMB UNDER IRRADIATION AND EXTERNAL STRESSES



α = Climb Distance Due to Irradiation Only
β = Climb Distance Due To Stress Only

10/72 FMT 2475

REDUCED IRRADIATION CREEP RATE FOR TORSION

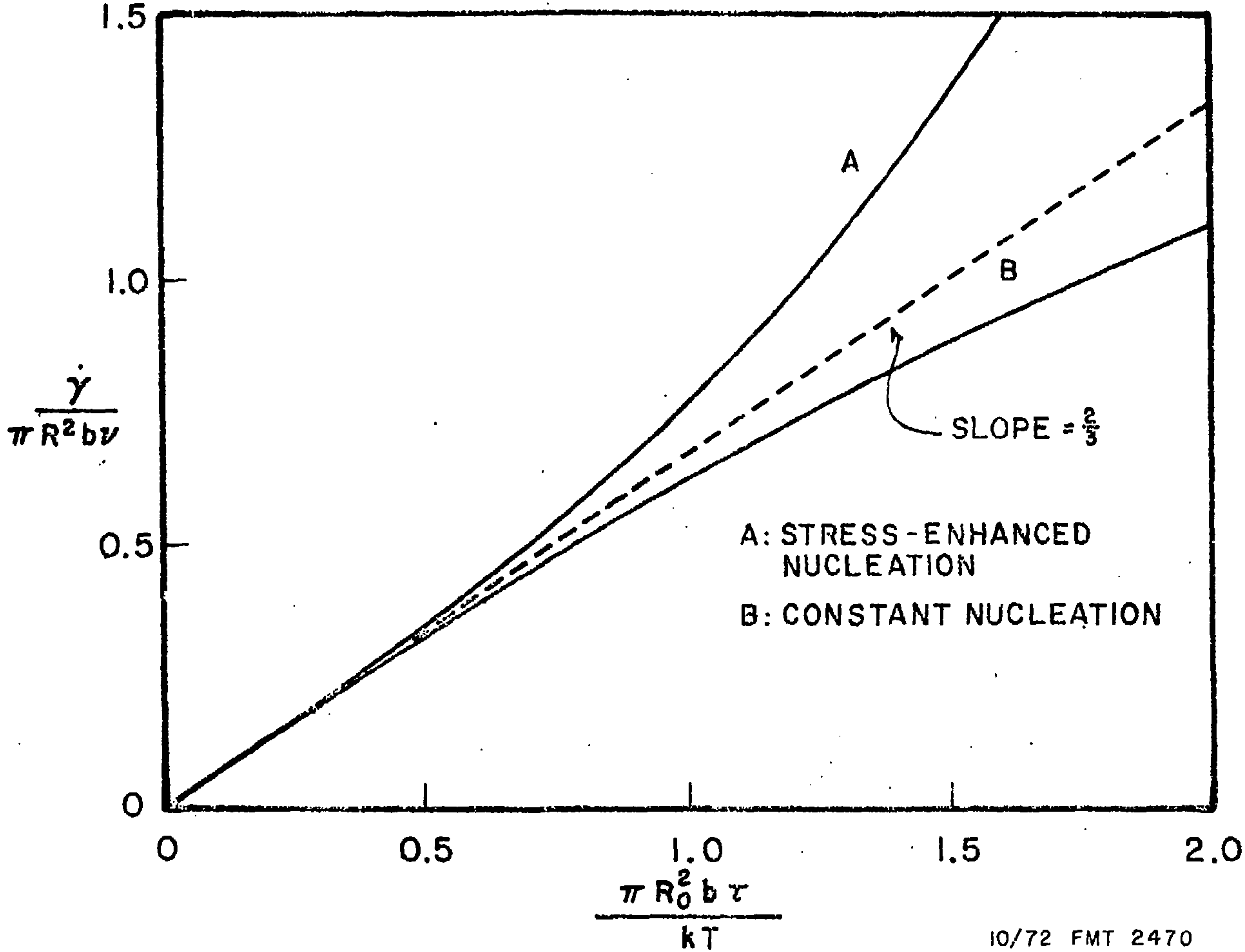


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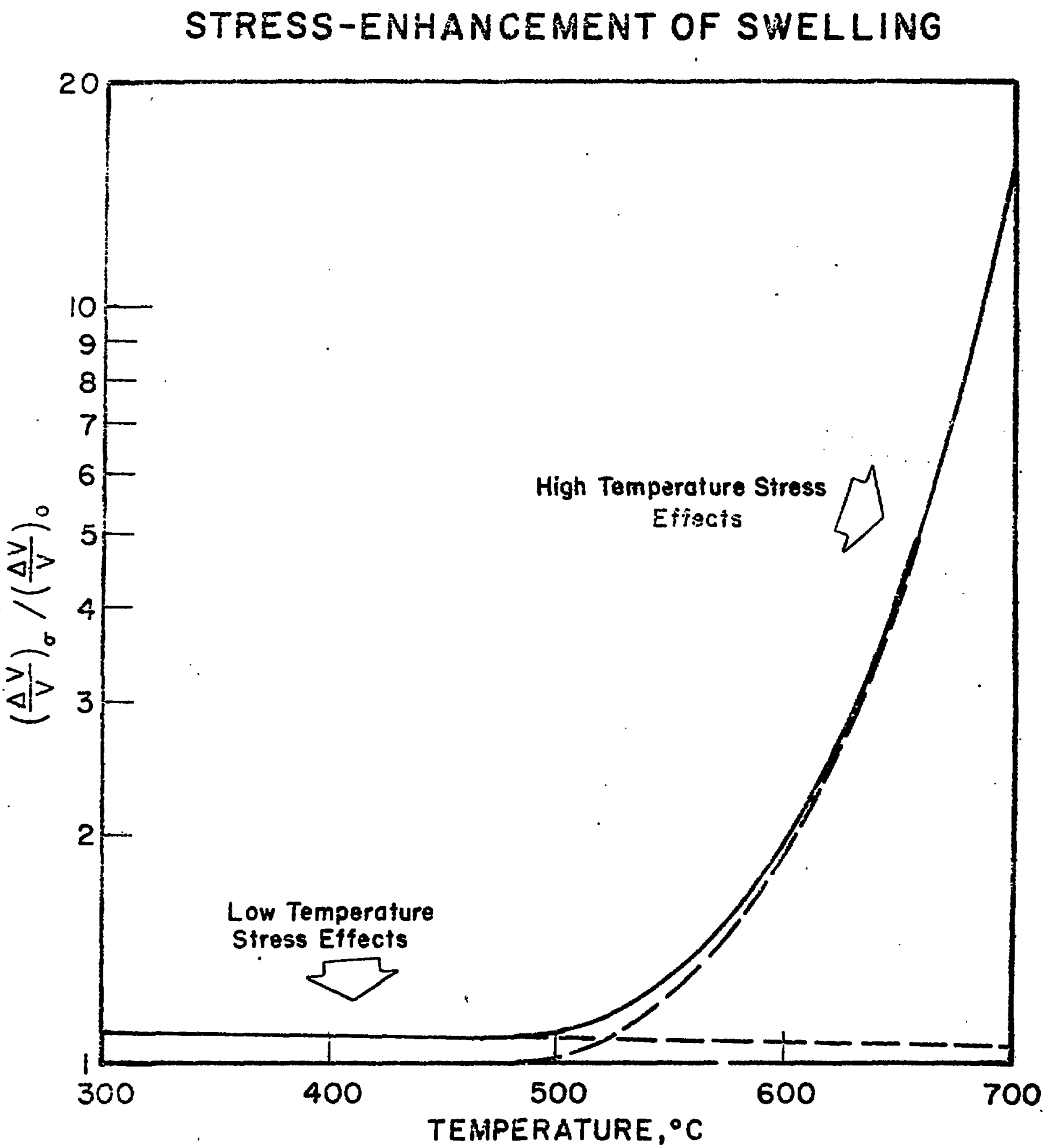
Figure 3



8/72 FMT 2421