

## IRRADIATION PARAMETERS INFLUENCED SWELLING OF STEELS

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### Abstract

Swelling of irradiated steels is generally affected by two categories of factors; microstructural defects and irradiation parameters. This paper studies the effects of irradiation parameters such as temperature, dose, and dose rate on the swelling of irradiated steels. The variation of percentage swelling with each of these parameters is first shown. An interpretation is then given in terms of microstructure parameters such as point defect concentrations, sink strengths, thermal vacancy emission, and point defects diffusion coefficients. It is found that the percentage swelling is not a monotonic function of temperature, the swelling has a similar shape for electron, ion or neutron-irradiation. It goes through a nonlinear then linear dose dependence regimes. In general increasing dose rate is found to be equivalent to a higher temperature than is actually employed.

## 1. Introduction

One of the most important parameters that affect swelling is temperature. Swelling occurs only in a limited temperature range and is peaking somewhere in that range. For stainless steel maximum swelling occurs at  $0.45 T_m$ , while void formation regime extends from  $0.3 T_m$  up to  $0.55 T_m$ . However, there are indications that some refractory metals, such as rhenium, niobium, molybdenum and titanium, develop voids, at rather lower temperature;  $-70.25 T_m$  (1).

The rate of change of the point defect concentration can be written as (2)

$$\frac{dC_i}{dt} = K_i - D_i C_i K_{it}^2 - \alpha C_i C_v \quad (1)$$

and

$$\frac{dC_v}{dt} = K_v - D_v C_v K_{vt}^2 - \alpha C_i C_v \quad (2)$$

where  $C_i$  and  $C_v$  are interstitial and vacancy concentrations in the matrix.  $D_i$ , and  $D_v$  are the diffusion coefficients for interstitials and vacancies, respectively.  $K_i$  and  $K_v$  are the effective point defect production rates,  $K_{il}^2$  and  $K_{vt}^2$  are the total sink strengths for interstitials and vacancies, respectively. At steady state, the rate of change of  $C_i$  and  $C_v$  is zero. Hence, equations 1 and 2 become the diffusion equations describing  $C_i$  and  $C_v$ .

At low temperatures, the mobility of point defects is low (diffusion coefficient is small). Therefore, the recombination term  $\alpha C_i C_v$  is dominating the two other terms in the diffusion equations 1 and 2. With few vacancies available for swelling, low swelling rate is expected at low temperatures.

After swelling has passed its peak value it decreases with increasing of temperature. The reduction in swelling at high temperature is due to the eventual dominance of the self-diffusion term in the equation.

$$\frac{dr_c}{dt} = -\frac{1}{b} [ D_v C_v^e - D_i C_i - D_v C_v^e \exp \left( -\frac{F_m \Omega}{KT} \right) ] \quad (3)$$

which describes the rate of change of void radius  $r_c$ . In this equations  $b$  is the Burger's vector,  $\Omega$  is the atomic volume,  $K$  is Boltzmann constant,  $T$  is the absolute temperature,  $F_m$  is the mechanical force tending to shrink the void, and  $C_v^e$  is the thermal equilibrium vacancy concentration. The dominance of self diffusion term tends to reduce vacancy supersaturation, hence, decrease the swelling.

A vital factor that affects swelling is the dislocation structure. An understanding of the temperature-dependence of irradiation-induced dislocation structure is desirable to aid that of void formation [1]. This is treated theoretically in this paper.

The effect of varying the displacement rate but keeping the total displacement dose constant is also considered in this work. This is of importance when comparing different types of irradiation. In general, a higher displacement rate is known to give results characteristic of

lower temperature than that actually employed [1], and vice versas. Packon [3] has found that the increase in neutron flux at constant fluence by a factor of 10 has increased the void concentration in aluminium by a factor of 2. The voids were smaller and there was no significant change in swelling.

The effect of displacement rate can be explained according to equations 1, 2 and 3. At low temperature, the recombination rate is dominating the rate of change of  $C_1$  and  $C_v$ . With increasing displacement rate, both  $C_v$  and  $C_1$  increase, resulting in greater recombination and consequently reduced void growth. At high temperature, the rate of change of void radius is controlled by the extent to which  $C_v$  exceeds the thermal equilibrium value  $C_v^e$ . Increasing the displacement rate increases  $C_v$  and consequently postpones the fall in swelling to a higher temperatures.

A typical empirical equation that expresses the swelling-fluence relationship is given by,

$$\frac{\Delta V}{V} \propto (\phi t)^n \quad (4)$$

where  $n$  is an exponent determined by best fit. It is about unity at 400°C and increases to about 2 at high temperatures. Other functional forms have been suggested for the fluence dependence of swelling. Because of the scatter of the data, swelling can equally well be fitted to a linear relation with an incubation period during which voids are absent [4],

$$\frac{\Delta V}{V} \propto \varphi t - (\varphi t)_0 \quad (5)$$

The incubation period  $(\varphi t)_0$ , is of the order of  $10^{22}$  n/cm<sup>2</sup> and is believed to represent the neutron dose needed to produce enough helium to permit void nucleation to proceed.

The incubation period may also be required to build up a sufficient density of interstitial loops to allow the preferential absorption of the interstitials by dislocations to sufficiently bias the point-defect population in the metal in favour of vacancies so as to permit vacancy agglomeration into voids.

In this paper the effects of temperature, irradiation dose rate, and total dose, on the swelling of type -316 stainless steel is investigated. Theritical treatment is based on the rate theory approach. Details of formulation, materials constants, and computer program used are shown in reference [5]. A computer code SWRAT is developed in which the temperature and dose-rate can be set to any value and swelling is calculated up to any dose at the set values.

## Results and Discussion

### 1. Temperature effect

Figure 1 shows the swelling-temperature relation for electron-irradiated 316 SS at a dose rate of  $5 \times 10^{-3}$  dpa/sec and dose of 40 dpa. It is clear that the relation is not monotonic. The interpretation presented earlier to this behavior is examined here. The mobility of vacancies with temperature is shown in Fig. 2 where vacancy and interstitial diffusivity-temperature variations are

plotted. Vacancy diffusion coefficient is very small at low temperature and increases rapidly. Interstitial diffusion coefficient is generally much higher than the vacancy diffusion coefficient. Accordingly, the small vacancy mobility at low temperatures leads to a large concentration of single vacancies. This ensures that the much more mobile interstitials have a chance of encountering a vacancy, and hence recombining instead of migrating to a sink.

At higher temperatures, the thermal equilibrium vacancy concentration becomes very high (Fig. 2). This produces high thermal emission rates. Irradiation-produced vacancies are lower in number than thermal vacancies. In other words the necessary vacancy supersaturation conditions for void growth is not prevailing in this temperature regime. The vacancy supersaturation  $S$  versus temperature is also shown in Fig. 2.

Figure 2 shows also clearly the two contradicting temperature sensitive parameters  $D_v$  and  $S$ . At low temperature the effect of the low  $D_v$  dominates that of high  $S$  since vacancies, although being at high concentrations, hardly migrate to voids. However at high temperatures the effect of low supersaturation dominates since the voids tend to evaporate rather than grow. At intermediate temperatures the two parameters compete and this results in a peak in the swelling at some intermediate temperature.

The interstitial loop dislocation density change with temperature is shown in Fig. 3. The reverse effect on swelling is not so direct. Increasing the dislocation loop density with decreasing temperature may affect the swelling in two different ways. As the interstitial loop density increases, the total dislocation density increases and

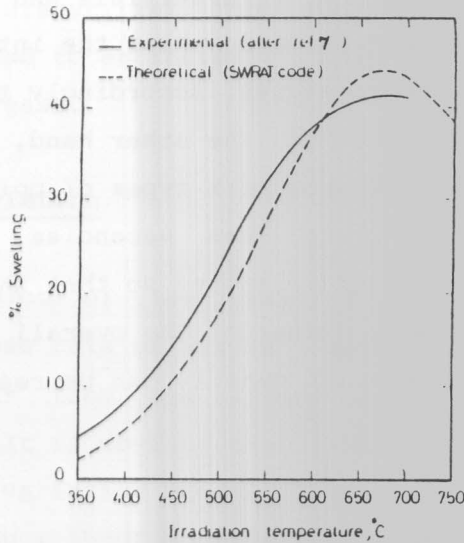


Figure 1: The temperature dependence of swelling in electron-irradiated 316 SS at a dose of 40 dpa and a dose rate of  $5 \times 10^{-3}$  dpa/sec.

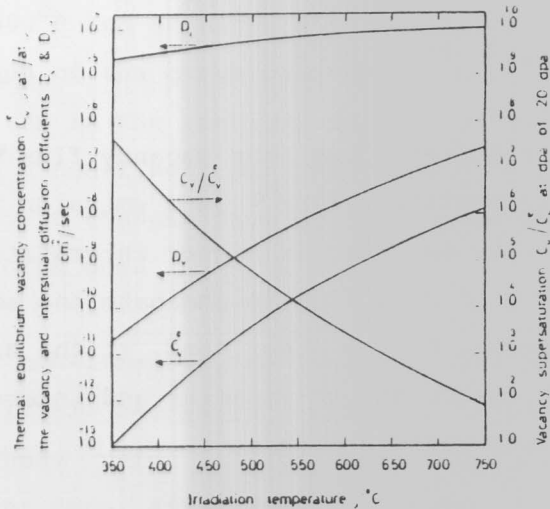


Figure 2: Thermal equilibrium vacancy concentration, diffusion coefficients and vacancy supersaturation versus temperature for electron-irradiated 316 SS.

accordingly the absorption of interstitials and vacancies increases. Due to the interstitial dislocation bias the interstitials are being absorbed more than the vacancies. Accordingly the total flux to the void enhances void growth. On the other hand, an interstitial loop represents a sink which absorbs both types of point defects. Although it absorbs interstitials more than vacancies, if it becomes large enough it would absorb enough vacancies so that the total vacancy flux will be affected by vacancy emission. The overall effect of increasing interstitial loop dislocation density can be realized by considering the void growth law given by,

$$\frac{dr_c}{dt} = \frac{1}{r_c} [\varphi_b - \varphi_e], \quad (6)$$

where  $\varphi_b$  is the bias flux and is given by,

$$\varphi_b = \varphi_v - \varphi_i, \quad (7)$$

and  $\varphi_e$  is the emission flux.

Thus the growth requires that the vacancy flux  $\varphi_v$  dominates the other two fluxes, interstitial flux  $\varphi_i$  and emission flux  $\varphi_e$ . If the dislocation density resulted in higher interstitial absorption and accordingly high enough bias flux to dominate the emission flux, void growth would increase. On the other hand, if the dislocation density is high enough to absorb enough vacancies and interstitials to reduce bias flux, void growth would decrease.

To summarize, from the presented discussion it can be shown that



increasing the interstitial loop density while decreasing the temperature would lead to an increase in the magnitude of the swelling followed by its decrease.

## 2. Swelling-Dose Variation

Fig. 4 shows a plot of the swelling versus dose for electron irradiation at a dose rate of  $5 \times 10^{-3}$  dpa/sec and a temperature of  $700^{\circ}\text{C}$ . It is clear from the figure that swelling continues to increase with dose. It is obvious that after a non-linear period, till about 15 dpa, the variation becomes linear. This is also the case in heavy-ion irradiation without considering vacancy loops which is shown in Figs. 5 and 6. It is suitable to mention here that changing the interstitial bias and/or the temperature, changes the start dose of the linear variation and changes the value of the swelling at the end of the non-linear period.

Figure 7 shows the swelling-dose variation for neutron irradiation at a temperature of  $600^{\circ}\text{C}$  and at different dose rates. In this case the change in the shape of the curve with the change in the dose rate, is easily observed. As in the previous cases, neutron irradiation shows the same trend of a non-linear part followed by a linear one. However the shape of the curve changes from gentle convex at low dose rates to gentle concave at high ones.

The change from non-linearity to linearity may be attributed to the difference in initial decreasing rates of vacancy and interstitial concentrations. These rates affect the flux rates of both point defects which, in turn, affect the void radius and consequently the swelling.

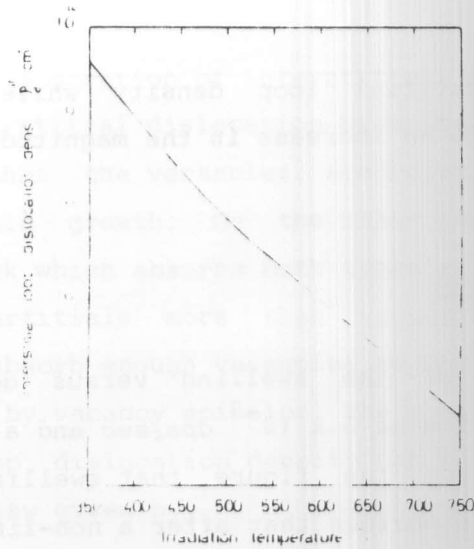


Figure 3: Interstitial loop dislocation density versus temperature for electron irradiation at a dose rate of  $5 \times 10^{-3}$  dpa/sec and a dose of 20 dpa.

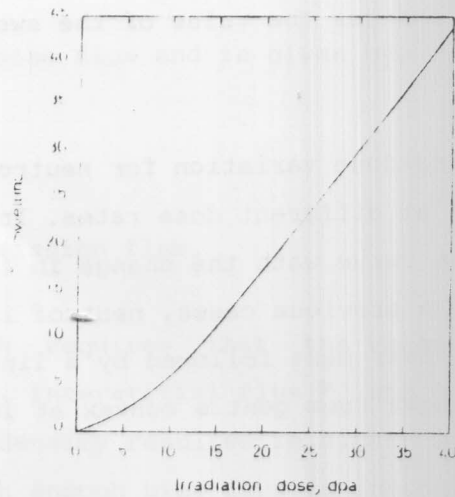


Figure 4: The swelling-dose variation for electron-irradiated 316 SS at a dose rate of  $5 \times 10^{-3}$  dpa/sec and a temperature of 700.

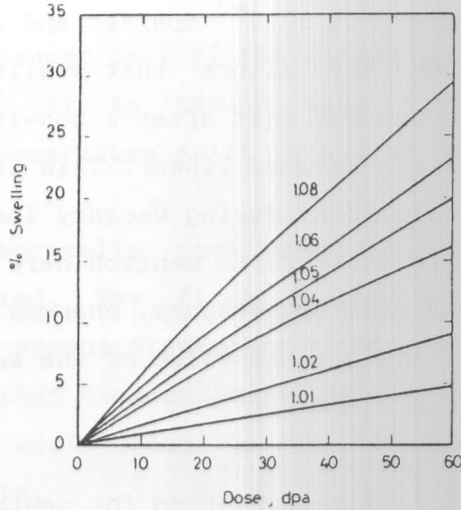


Figure 5: Swelling versus displacement dose for ion-irradiated 316 SS at a dose rate of  $1 \times 10^{-3}$  dpa/sec and at temperature of  $400^\circ\text{C}$  for different values of interstitial bias.

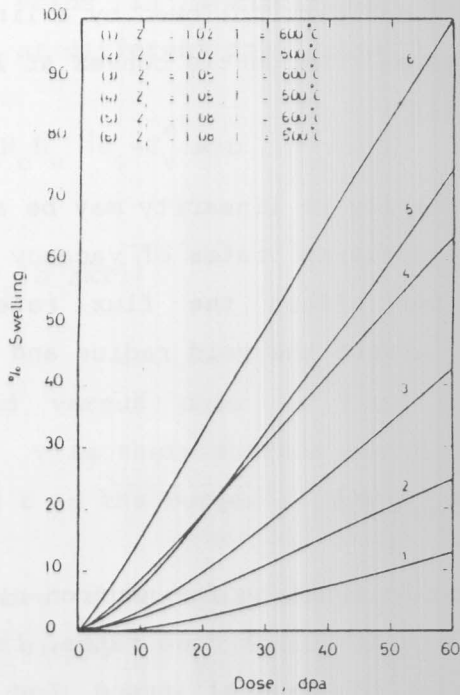


Figure 6: Swelling versus displacement dose for ion-irradiated 316 SS at a dose rate of  $1 \times 10^{-3}$  dpa/sec at different values of the interstitial bias at different temperatures.

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The change from non-linearity to linearity may be attributed to the difference in initial decreasing rates of vacancy and interstitial concentrations. These rates affect the flux rates of both point defects which, in turn, affect the void radius and consequently the swelling.

### 3. The Effect Of Dose Rate (Flux)

Fig. 7 shows the variation in swelling for neutron-irradiated 316 SS at a temperature of  $600^{\circ}\text{C}$  and different dose rates. It is clear from the figure that there is no general trend for the behaviour of swelling with dose rate except what we have mentioned before about the

shape of the nonlinear part of the curves which reflects the initial void growth rate. It is thought that increasing the dose rate corresponds to a temperature shift to higher values. [1,6]

To show that numerically some important parameters that affect the process are plotted. The first is the vacancy concentration  $C_v$ , which is plotted versus displacement dose at different dose rates in Fig. 8. It shows that vacancy concentration increases with increasing dose rate. This would lead to an increase in recombination with increasing dose rate.

Another very important factor which is a measure of the vacancy absorption by void is  $Q_v^{void}$ , the rate of absorption of vacancies by all of the voids in a unit volume of solid, is plotted versus displacement dose at different dose rates.  $Q_v^{void}$  is given by,

$$Q_v^{void} = 4 \pi r_c N_c D_v \left\{ C_v - C_v^e \exp \left[ \left( \frac{2\gamma}{r_c} - P_g \right) b^3 / KT \right] \right\} \quad (8)$$

which is plotted versus dose in Fig. 9, where  $N_c$  is the number density of voids,  $\gamma$  is their surface energy,  $P_g$  is their internal gas pressure, and  $b$  is the Burger's vector.

Vacancies are more effectively absorbed at higher displacement dose rates. This may simply be attributed to high point defect production rates at high dose rates. Fig. 10 shows the ratio between the absorption rate of vacancies by the voids and the loss rate of both vacancies and interstitials by bulk recombination versus dose. This

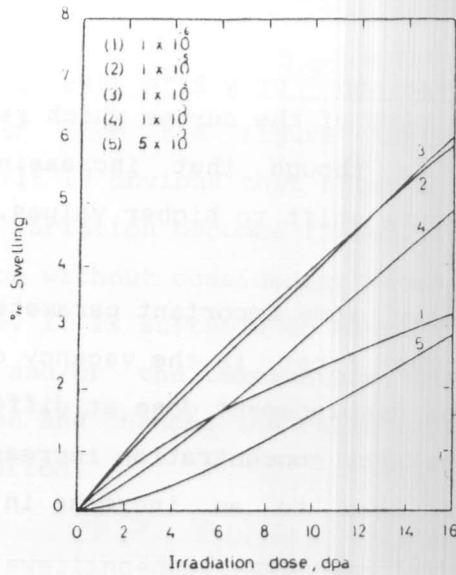


Figure 7: The swelling-dose variation for neutron irradiated 316 SS at temperature of 600°C and different dose rates (dpa/sec).

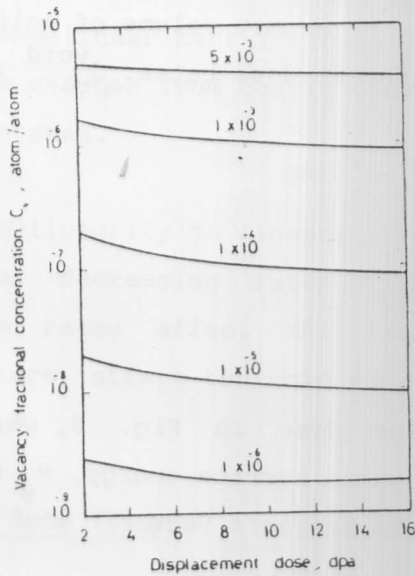


Figure 8: Vacancy fractional concentration versus displacement dose for neutron-irradiated 316 SS at a temperature of 500°C for different dose rates.

curve reflects the sharing ratio of both mechanisms to vacancy removal of the matrix. The dotted line in Fig. 10 is the line at which both mechanisms remove vacancies from the medium at equal rates.

One can easily conclude that at high dose rates recombination dominates absorption by voids. At low dose rates, the rate of vacancy absorption by voids is much higher than the loss due to bulk recombination. The higher recombination at high dose rates assures the idea of swelling shift to higher temperatures. As we have already mentioned, the high recombination is encountered also at low temperatures. The two final results of a high dose rate and a low temperature are similar despite the different mechanisms. As the higher recombination at high dose rates is attributed to high concentrations due to high production rates of point defects, the high recombination at low temperature is attributed to low mobility of vacancies so that they recombine with the more mobile interstitials. Fig. 11 shows an example of the increase of recombination during temperature decrease. Again, this shows that the idea of swelling shift to higher temperatures is equivalent to increasing the dose rate.

### Conclusion

Swelling of type 316 stainless steel is influenced by irradiation parameters such as temperature, dose, and dose rate through the changes in vacancy and interstitial concentrations. Swelling is not observed at low temperatures because of the very low mobility of vacancies. On the other hand, at high temperatures thermal emission of vacancies is high and voids evaporate rather than grow. These two processes oppose each other at intermediate temperatures and swelling

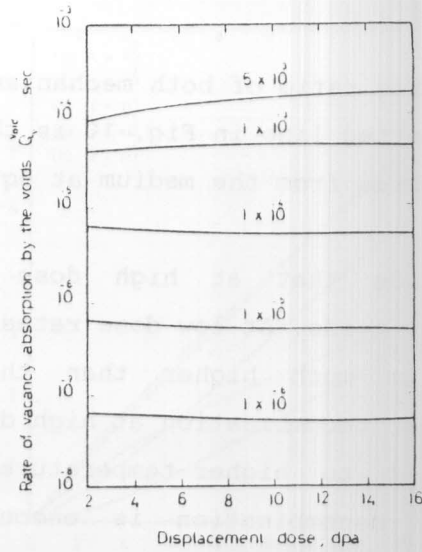


Figure 9: The rate of absorption of vacancies by all of the voids in a unit volume of solid versus dose for neutron-irradiated 316 SS at a temperature of 600°C and at different dose rates.

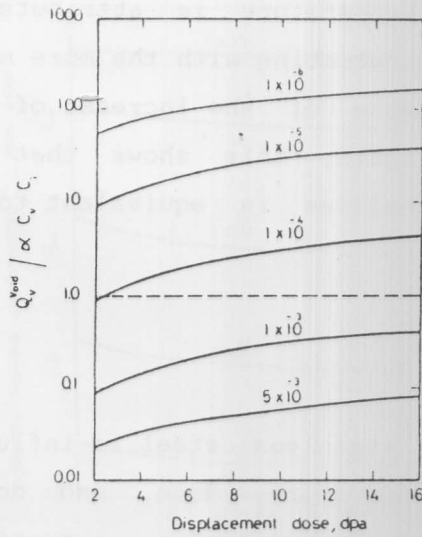


Figure 10: Ratio between absorption rate of vacancies by the void and loss rate of both vacancies and interstitials by bulk recombination versus dose for neutron irradiation at different dose rates.



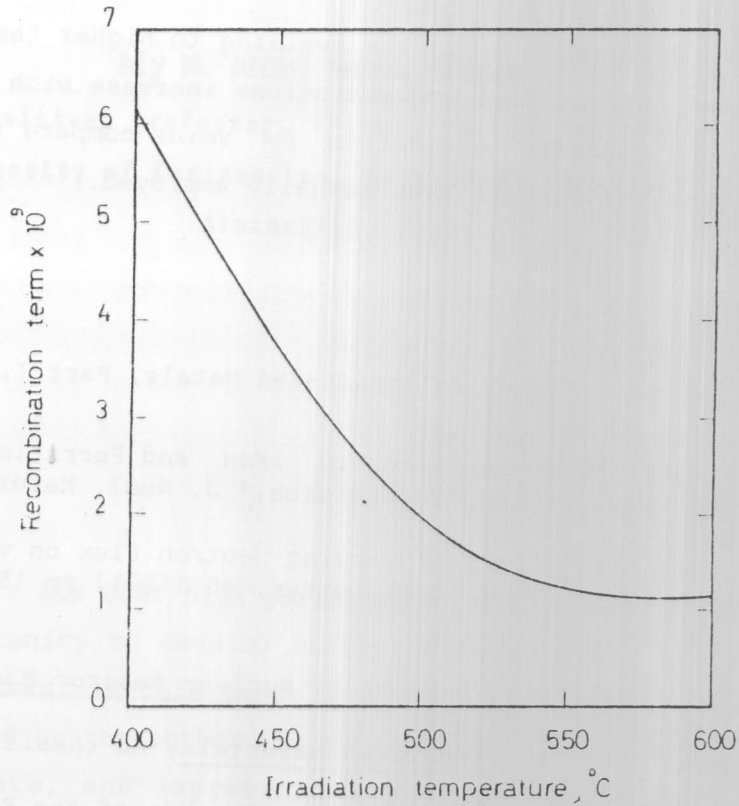


Figure 11 : The rate of loss of both vacancies and interstitials by bulk recombination versus irradiation temperature for neutron irradiation at a dose of 2 dpa and a dose rate of  $1 \times 10^5$  dpa/sec.

reaches a maximum value.

The swelling-dose relation passes through a nonlinear then a linear regimes. The shape of the non-linear part depends on the current values of various parameters such as temperature, dose, and microstructural composition.

Increasing the dose rate shifts the swelling to higher temperatures. While vacancy and interstitial concentrations increase with dose rate, recombination and vacancy absorption by voids compete to show the effect of a higher temperature than actually employed.

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