# A computer simulation of the low temperature neutron irradiation, high temperature annealing mode of void formation in molybdenum

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# A Computer Simulation of the Low Temperature Neutron Irradiation, High Temperature Annealing Mode of Void Formation in Molybdenum

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pages + tables + illustrations

**Abstract**

A computer model has been developed to simulate the processes leading to void growth during the post-irradiation annealing of low-temperature neutron irradiated molybdenum. The results appear to confirm that the model of void formation previously proposed could lead to the experimentally determined high efficiency of vacancy retention and give swelling rates one or two orders greater than that possible in high temperature void formation. Some of the important parameters are identified and discussed. There appeared to be no reason why the irradiation/annealing mode of void formation should not apply to metals other than molybdenum.

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## 1. Introduction

During an investigation in 1972 of the substructure of molybdenum neutron irradiated at 50°C to a dose of $5 \times 10^{19}$ n/cm$^2$ and subsequently annealed to 900°C, it was found by using electron microscopy that in certain circumstances voids could be formed[1]. The fact that in a ten year history of similar experiments voids had not previously been seen made the result a surprising one. The authors suggested that only in certain cases involving the presence of gas atoms could voids nucleate; the unpredictability of their own results - some specimens producing no voids - emphasised this point. In a subsequent note the present author[2] concentrated on the swelling produced by the voids and came to the unexpected conclusion that the efficiency of the void formation (in terms of the ratio of vacancies retained in voids to the number created in the irradiation) was some fifty times larger than even the maximum possible in the high temperature 'preference' mechanism of void formation. It was therefore necessary to put forward a different mechanism. This postulated that during the low temperature irradiation the vacancies in the displacement cascades either collapsed as vacancy loops or, if the nucleation conditions were right, as voids while a considerable fraction of the interstitials were essentially removed from the system by being trapped at interstitial loops. On annealing it was suggested that the vacancy loops shrank by thermal evaporation and fed vacancies into the voids thus enabling the voids to grow.

The model was based to a considerable extent on the results of a positron annihilation study[3] of the processes taking place during the post irradiation annealing of molybdenum which, as in the microscopy work, had been neutron irradiated at 50°C.

Although further experimental studies have been initiated, particularly to clarify the nucleation mechanism, it was felt that something could be gained by carrying out a computer simulation of the neutron irradiation and post-irradiation annealing processes on the basis of the proposed void growth mechanism. In this way it was hoped to identify the important parameters and check whether the model could give results of the same order as those obtained experimentally[1] where a void swelling of 0.36% indicated a value of 14% for the efficiency of vacancy retention.

The outline of the model is given in more detail in section 2, the results in section 3 while aspects of both are considered in the discussion section.

## 2. Computer Model

The model of the irradiation and annealing processes can be split into five main sections as follows, the first two for the irradiation and the remaining three for the annealing.

(i) The distribution of defects in the collision cascade.

(ii) The migration and trapping or annihilation of the mobile interstitials produced in the cascade event.

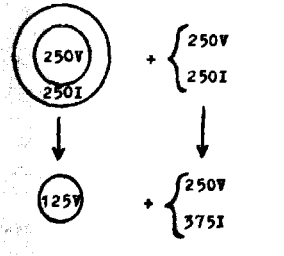
(iii) The migration of vacancies at the start of the post-irradiation annealing and their distribution to sinks in the system.

(iv) The thermal evaporation of vacancies from vacancy loops during annealing and their distribution to sinks in the system, i.e. voids and loops.

(v) The coalescence of interstitial loops by glide and climb processes.

These five main stages are indicated in the overall simplified flow diagram in figure 1 and we shall discuss each of them in turn. Some smaller considerations are discussed in section (vi).

(i) The complexity of the collision cascade is self-evident and it is necessary at this stage to introduce fairly severe approximations. The overall picture of an average cascade is one in which a central spherical vacancy-rich zone is surrounded by a shell of interstitials while single interstitials and vacancies are also created away from this 'spike' region. We have chosen to represent the initial stage of cascade formation as one (see inset) in which the central core contains 250 vacancies surrounded by 250 interstitials while an additional 250 vacancies and interstitials are created randomly in the lattice. In the second stage of cascade formation the mobile interstitials must cause considerable recombination within the cascade; this was taken into account by assuming that half the interstitials migrate inwards and half outwards. The overall result can be seen in the inset with a core of 125 vacancies in the cascade and 250 vacancies and 375 interstitials in the lattice. In the final stage of the collision cascade we consider what






happens to the core of vacancies. Although for irradiations below stage 3 (0.15 Tm) the vacancies are immobile it is thought that there is easily sufficient thermal energy in the cascade region for the vacancies there to migrate and collapse either to vacancy loops or, if nucleation conditions allow, to voids.

In order to study the effect of different amounts of void nucleation an adjustable parameter was included to control the fraction of cascades collapsing as voids.

(ii) The next event in the irradiation sequence was the migration of the free interstitials to the sinks in the system, i.e. to the voids or vacancy loops created in the cascades and to the free immobile vacancies, where the interstitials are annihilated, and to other sinks where the interstitials are trapped. Initially in the real case these latter sinks are impurity atoms which then by the acquisition of interstitials become interstitial lopps. In the program we start with a given adjustable concentration of interstitial loops of one Burgers vector radius.

The fraction of interstitials going to each of the sinks was determined by the cross-section (see (vi)) of the sink. Clearly the sum of cross-sections for the voids, vacancy loops and vacancies could be termed the annihilation cross-section while the total cross-section of the interstitial loops could be called the trapping cross-section. The value of these individual cross-sections (determined by the number of defects each contained) was continuously calculated by the computer as each sink component lost or gained a defect.

As shown in figure 1 the irradiation consisted of a repetition of (i) and (ii), one cascade per million atoms being put in on each cycle. During the irradiation therefore, as successive cascades were created an array of voids and vacancy loops was built up together with a population of free vacancies. Although these voids/loops and vacancies are continually eroded by the interstitials - for example the voids/loops formed early in the irradiation will eventually shrink completely by this process - provided interstitials continue to be trapped or interstitial loops then the overall number of vacancies must continually increase during the irradiation. Because of the trapping of interstitials the concentration of interstitial loop nuclei becomes an extremely important parameter in the model.

(iii) The migration of free vacancies will not occur during the irradiation provided the temperature is sufficiently low. However on annealing there is fairly substantial evidence that the vacancies in molybdenum migrate in

the so-called stage 3 region occurring at around 200°C. The simulation of this is exactly along the lines of the interstitial migration during irradiation, that is the vacancies are distributed to the various sinks in the system according to their cross-section, either adding to the voids and vacancy loops or shrinking the interstitial loops.

The possibility that voids could nucleate at this point, by the capture of vacancies or gas atoms, is discussed in section (4). It was simple to include this in the program.

(iv) An important part of the model of void growth during annealing is the fact that vacancy loops will shrink by thermal evaporation of vacancies and thus cause vacancies to be transferred to the voids. The basic equation for vacancy loop shrinkage is

$$\frac{dr}{dt} = A\,Do\,\exp(-Q/kT)\left[\frac{c}{co} - \exp\left(\frac{\tau b^2}{rkT}\right)\right] \qquad (1)$$

where A is a geometrical factor equal to $\left(\frac{4\sqrt{3}}{a}\right)$ where a is the lattice parameter; Do, Q, k and T have their usual meanings; Co is the equilibrium vacancy concentration appropriate to temperature T; c is the actual vacancy concentration; $\tau$ is the line tension of the loop, b its Burgers vector and r its radius.

The second term represents the evaporation of vacancies while the c/co term represents the condensation of vacancies onto the loop. To overcome the difficulties in calculating the actual value of c the following procedure was used: first, by considering only the evaporation term in equation (1), (dr/dt) was calculated for each vacancy loop in the system and the total number of vacancies released to the lattice computed. The condensation of these vacancies was then carried out by distributing them to the sinks in the system according to the cross-section of each sink. In this way some vacancies returned to the vacancy loops (in accordance with the c/co term) while others were acquired by voids or annihilated at interstitial loops. As expected from the 1/r term in the exponential the small vacancy loops shrank faster than the large ones and in fact the larger loops grew until the temperature increased sufficiently for the evaporation/shrinkage term to dominate. All the vacancy loops eventually shrank and released their vacancies to the system where they either fed the voids present or were annihilated at the interstitial loops.



(v) There are many experiments demonstrating that the annealing of irradiated structures causes a rearrangement of the dislocation structures, a considerable coarsening taking place as the annealing temperature is raised in the range ~ 400 to 700°C[4, 5]. From the interstitial loop point of view there are two mechanisms involved; firstly the loops can coalesce by glide processes, or secondly they can climb by the short circuit diffusion of vacancies around the loop periphery. The mechanisms have been discussed in detail by, for example, Eyre and Maher[4] and by Brinhall and Mastell[5].

The importance of coalescence in the present model lies in the fact that for a given number of interstitials in the system their effectiveness as sinks for vacancies is a maximum when they are distributed in the form of many small loops rather than a few large ones. Conversely then, when coalescence takes place their sink cross-section drops and the net effect is that the vacancies released from vacancy loops have a far greater chance of ultimately being trapped at voids rather than being annihilated at interstitial loops.

The main problem, however, is how to take the coalescence into account in an adequate fashion since the real situation with its variety of loop sizes and orientations is a highly complex one. We have used an approach based on that developed by Eyre and Maher[4]. They demonstrated that their experimental results on the annealing of loops in molybdenum were consistent with their treatment of the glide and climb mechanisms. The centre of their approach is the derivation of two critical spacing values for glide and climb respectively which are then combined to define a critical volume. If the average volume per loop is less than the critical volume (i.e. the loop density is larger than the critical loop density = 1/critical volume) then coalescence takes place so as to increase the volume of crystal available to each loop, i.e. to increase the interloop spacing.

The derived value for the critical volume Vc is given by the expression

$$Vc = 2\pi \sin 32^\circ \; Rg \cdot Rc^2$$

where Rg and Rc, the critical spacings for glide and climb respectively are given by

$$Rg = \left[1.41\,\frac{\mu b^2}{4(1-\nu)} \cdot \frac{r^2}{\sigma g}\right]^{1/4}$$

$$Rc = \left[5\,\frac{\mu b^2}{4(1-\nu)} \cdot t \cdot a^5\,\frac{v_o r}{2kT}\exp(-Q_L/kT) \cdot 3\right]^{1/5}$$

where $\mu$ = bulk shear modulus, $1.23 \cdot 10^{12}$ dynes/cm$^2$; b = Burgers vector; 2.78 Å; t = annealing time; a = lattice parameter, 3.146 Å; $\nu_o$ = frequency factor, $10^{13}$; r = loop radius; $Q_L$ = activation energy for pipe diffusion, 1.72 eV; $\sigma_g$ = tensile yield stress, $5.10^7$ dynes/cm$^2$; $\nu$ = Poissons ratio, 0.385.

These values lead to the following expression for Vc

$$Vc = 1.15 \cdot 10^{10} \left[\exp(-2.0 \cdot 10^4/T) \cdot t/T\right]^{2/5} \cdot r^{0.9} \ (\text{Å})^3$$

and we have used this equation in the computer model. The main characteristic of the equation is that Vc increases with temperature and annealing time so that during an isochronal anneal where the specimen is subject to linear heating a temperature will eventually be reached where, for a given interstitial loop density and loop radius, Vc will exceed the actual average volume per loop. Coalescence will then take place. In the program where all the interstitial loops are the same size this is carried out by lowering the loop density and increasing their radius so as to leave the total number of interstitials in the loops unchanged.

(vi) We include here some smaller considerations.

(a) Cross-sections. The importance of the sink cross-sections in the present model is self-evident, the relative value of the cross-section of any particular sink determining the fraction it will capture of those defects mobile at that time. The cross-section of the voids and loops (vacancy and interstitial) were taken as $4\pi \cdot$ radius and $2\pi \cdot$ radius respectively[6,7] while the single vacancies were assumed to be equivilant to voids of diameter equal to the Burgers vector.

(b) Annealing. Starting at 300°C the temperature was increased by degree intervals, each temperature being held constant for a given time. At each temperature the vacancy loop shrinkage was calculated and the coalescence of interstitial loops considered. The program stopped when all the vacancy loops had shrunk though if required the annealing of the voids could also have been considered.

(c) Irradiation Dose. In the model the doses are expressed in displacements per atom, 500 displacements per million atom being introduced on each cycle. For convenience however we have converted the results in section 3 to neutrons/cm$^2$ on the basis of $10^{19}$ n/cm$^2$ being equal to 0.005 displacements per atom.



## 3. Results

We first examine the retention of defects during the irradiation. The parameter of interest here is the amount of interstitial loop nucleation which has been varied from 0.1 to 100 parts per million (p.p.m.). The large effect of this parameter is illustrated in figure 2 where the efficiency of defect retention is plotted against dose. Since the efficiency is the percentage of the created defects that are retained in the lattice it is worth recalling that in the model considered, 25% of the created defects are annihilated immediately so that the theoretical maximum is only 75% and not 100%. An alternative method of presenting the data in figure 2 is to plot the efficiency against interstitial loop concentration; we show this in figure 3. In addition the dotted line gives the efficiency for the $5 \times 10^{19}$ n/cm$^2$ dose after stage 3 annealing, i.e. after the free vacancies have been distributed to sinks.

Since the amount of defect retention during irradiation is controlled basically by the probability of the migrating interstitials being annihilated rather than being trapped, it is of interest to examine the variation of this probability with dose and interstitial loop concentration as shown in figure 4. Again the large effect of interstitial loop nucleation is illustrated. One point to note is that during the migration of vacancies in stage 3, the ratio in figure 4 is inversed and becomes the ratio of trapping to annihilation. An effect of this is that the loss of defects in stage 3 increases with interstitial loop concentration in contrast with the reverse situation during irradiation. The amount of drop in figure 3 from the $5 \times 10^{19}$ line to the dotted line illustrates this effect.

We now go on to examine the vacancy retention after thermally annealing out all the vacancy loops and redistributing the released vacancies to the other sinks in the system, i.e. voids and interstitial loops (taking into account their coalescence). An important parameter now is the percentage of the original cascades that have collapsed as voids rather than vacancy loops. The effect of this can be seen in figure 5 where, after an irradiation dose of $5 \times 10^{19}$ n/cm$^2$, and after stage 3 and vacancy loop annealing, the efficiency is plotted as a function of original interstitial loop concentration and the void nucleation percentage. The important result, relevant the experimental value of 14% vacancy retention after the same irradiation dose, is the fact that efficiencies of this order can be comfortably attained for values of interstitial loop nucleation greater than 10 p.p.m. and even with only 10% of the cascades nucleating as voids.

At this stage it is worth recalling the fact that for the high temperature mode of void formation the maximum possible efficiency is one-quarter the preference[8], i.e. between 1/4 and 1/2% for preference values between 1 and 2%. Thus the swelling figures in figure 5 for a dose of $5 \times 10^{19}$ neutron/$cm^2$ (or .025 dpa) could be compared to the maximum swelling on the preference mechanism of between .006 and .012%. Although the two mechanisms apply in different circumstances the comparison is still of some interest.

One point emerges from the results concerning the amount of void nucleation; this is simply that the final efficiency is far from being linearly proportional to the nucleation. In other words variations in void nucleation will not grossly affect the final efficiency values. The real reason for this lies in the interstitial loop coalescence since once the interstitial loop cross-section becomes small compared to the void and vacancy loop cross-sections then the fraction of vacancies lost in the system by annihilation also becomes small and the dependence of the efficiency on the number of void nuclei is clearly diminished. The processes occuring during annealing can be followed in figures 6 and 7, where various parameters are plotted against annealing temperature after a dose of $5 \times 10^{19}$ n/$cm^2$ and for a representative case of 10 p.p.m. interstitial loop nucleation and 50% void nucleation in the cascades (giving an equal number of vacancy loops and voids). Figure 6 shows the capture cross-sections for the vacancy loops, voids and interstitial loops as a function of temperature and illustrates several important points: (1) the drop in vacancy loop cross-section as the vacancy loops shrink; (2) the rise in void cross-section as the voids act as sinks for the vacancies released from vacancy loops; (3) the sharp drop in interstitial loop cross-section as the loops coalesce. The importance of this coalescence is emphasised by the plot in figure 7 of the ratio of trapping to annihilation cross-sections, for the vacancy, as a function of temperature. Once coalescence begins - in this case at 600°C - there is a continuous rise in the probability that a vacancy evaporated from a vacancy loop is either recaptured at a vacancy loop or reaches a void rather than being annihilated at an interstitial loop. The final predominant process is then the simple transfer of vacancies from the vacancy loops to the voids. The plot of actual defect numbers in figure 7 illustrates this point.

In computer programs where interstitial coalescence was not included the computed efficiences were of the order of one-quarter of the values shown in figure 5.



The temperature, Tf, at which the vacancy loops completely disappear is worth discussing briefly in terms of the variable parameters. The interstitial loop concentration has a relatively small effect, the value of Tf increasing by about 50°C as the concentration changes from 1 p.p.m. to 100 p.p.m. The amount of void nucleation has a far greater effect; relative to 50% nucleation, 10% void nucleation increases Tf by 150°C and reflects the fact that because only a few voids are present a larger fraction of vacancies released from the vacancy loops are recaptured. The overall shrinkage rate of the loops is then very much reduced and they survive to higher temperatures. One other parameter is worth notice, that of the isochronal heating rate, $\partial T/\partial t$. The rate used in all the calculations has normally been 1°C/second (i.e. 300°C to 900°C in 10 minutes) but in figure 6 the dotted lines refer to the far slower rate of 1°C/minute. This has a marked effect both on the temperature of interstitial loop coalescence and on the vacancy loop shrinkage. The final Tf now in fact comes in the range of that found by Petersen et al.[3] in their positron annihilation study where such a slow heating rate was used. However it must be stated that there are too many unknown parameters - such as the vacancy diffusivity, the self-diffusion energy and the line tension to predict Tf accurately.

## 4. Discussion

Although it is clear that a simulation of the processes as complex as that of neutron irradiation and annealing must involve numerous simplifications and assumptions, together with the use of many adjustable variables, it is felt that the computer simulation has demonstrated that the model put forward for the irradiation mode of void formation could explain the high efficiency of vacancy retention - and the associated high void swelling rate - found in the experimental results[1]. Furthermore the important parameters and processes have been identified. Among these are (i) the interstitial loop nucleation during irradiation, (ii) the amount of void nucleation and (iii) the interstitial loop coalescence:

(i) The effect of the interstitial loop concentration on the trapping of the interstitials created during irradiation, and therefore on the fraction of defects retained, is clearly shown in the results section. One problem however is to know what values of loop concentration might be expected in practical circumstances but fortunately some idea can be obtained from electron irradiation studies. In these the accepted model to explain damage rate curves between stages I and III involves the trapping of mobile inter-

stitials at impurity sinks and this is clearly the basis of the model put forward in the present paper for the neutron irradiation case. The advantage of electron irradiation studies are their relative simplicity which make it possible to extract information about the parameters involved. In such a study[9] of resistivity increment as a function of electron dose the present author found on two specimens of zone refined molybdenum - of resistivity ratios 5000 and 2000 - that the interstitial sink concentration was 3.5 and 9.5 p.p.m. respectively. For commercial purity molybdenum, as used in the experimental study, interstitial nuclei concentrations well above 10 p.p.m. might then easily be expected. Figure 5 in the results section shows that for this range there is no difficulty in reproducing the experimental result with the theoretical model.

(ii) One of the experimental facts regarding the annealing of neutron irradiated molybdenum is the rarity of void formation. This must point to some particular difficulty concerning void nucleation. In the model outlined only one nucleation situation has been considered, that of nucleation in the cascade, but it is not impossible that nucleation could take place at a later stage - during the migration of the free vacancies for example or during the release of vacancies from vacancy loops. Computer runs for the situation where nucleation took place in stage 3 in fact gave comparable results (for the same void concentrations) as for nucleation in the cascade.

Leaving this possibility aside for the moment, it is worth briefly examining the probability of void nucleation in the cascade. Assuming that gas atoms within an uncollapsed cascade are a prerequisite for the cascade to collapse as a void, and taking the extreme case that only one gas atom is required, then, if the thermal spike covers 1000 atomic columes the concentration of gas atoms for 100% void nucleation must be 1 in 1000; for 10% void nucleation it must be 1/10 of this, i.e. 100 p.p.m. If, as is likely, more than one gas atom is needed then it can be seen that the required concentration of gas atoms quickly becomes comparatively large.

One intersting point arises from this simple consideration of void nucleation in the cascade; this is, that if more than one gas atom is necessary for nucleation then it is better for the required atoms to be clustered together rather than be randomly placed individually in the lattice. Consider the situation where say 10 gas atoms are required; if the gas atoms are at random then 100% void nucleation will take place if there are 10 gas atoms per cascade volume, i.e. $10^4$ p.p.m. However for 10% nucleation one cannot say that a gas atom concentration of $10^3$ p.p.m. will suffice - it



clearly will not if the gas atoms are at random for the chances of any 1000 atoms having 10 gas atoms is remote. However if the gas atoms are already clustered in groups of 10 then the proportionality will hold as for the single atom case. It might therefore be far better from the point of view of void nucleation to have small clusters of gas atoms.

In any case it seems clear that the required concentrations of gas atoms mentioned above must make the possibility of void nucleation very remote in normal cases and this explains the rarity of void production by the irradiation/annealing method. In the only two sets of work where voids were found it is suggested that the pre-irradiation anneals could have led to gaseous contamination sufficient to allow void nucleation.

Returning to the case of void nucleation during the post-irradiation annealing an experiment was designed to try to test this possibility. The efficacy of helium as a nucleating agent is well substantiated so some of the neutron irradiated samples from previous work[1] were injected with a concentration of $10^{-5}$ helium ions and then annealed up to 900°C. In spite of a very thorough examination no voids were seen; in fact the dislocation substructure consisted of a high density of small loops characteristic of the non-void case. It is difficult to ascertain exactly why the helium failed to nucleate voids but the result can be taken to substantiate the idea of nucleation in the cascade rather than after the irradiation. In any case the result again illustrates the difficulty of getting the correct conditions for void nucleation.

(iii) The addition of interstitial loop coalescence to the annealing processes has a marked effect on the computed efficiency values. Although the coalescence part of the program contains the greatest simplification between the real and the computer cases, it is not felt that this simplification is of too much consequence. The exact temperature and speed of coalescence is not vitally important to the computed result provided that the coalescence starts before the large scale vacancy evaporation from the vacancy loops. This is an aspect that can only be determined experimentally but the sparse evidence is that this condition is met.

One interesting experimental result, which is explained by the model, is the different behaviour of the interstitial component in the presence or absence of voids. If voids are present the interstitial substructure develops into a coarse dislocation network by 900°C, but in the absence of voids the development is markedly slower giving a substructure of a high density of interstitial loops[1]. The reason for the difference is that if no voids are in the system then the interstitial loops are the only sinks for the vacancies

released by the vacancy loops. The interstitial loops therefore shrink and can keep the average loop volume above the critical volume for coalescence. On the other hand if voids are present and also collect vacancies, the interstitial loop shrinkage will be smaller and the critical volume conditions more easily met. Furthermore once coalescence does start, the fraction of further vacancies acquired drops, hence making further coalescence - as the temperature is raised - more easy and eventually a dislocation network is formed.

Finally there are two further short but important points which might point the way to future experimental work. Firstly there appears to be no reason why the irradiation/annealing mechanism of void formation should not be found on metals other than molybdenum. Secondly in the present study no reasons were found to disagree with the suggestion put forward previously[2] that the mechanism could operate for irradiation temperatures up to the vacancy loop shrinkage regime (~0.3 Tm). Future computer work will examine this aspect and particularly test the suggestion that thermal cycling about 0.3 Tm might cause void swelling at a rate considerably in excess of that predicted by the high temperature preference model.

## Acknowledgement

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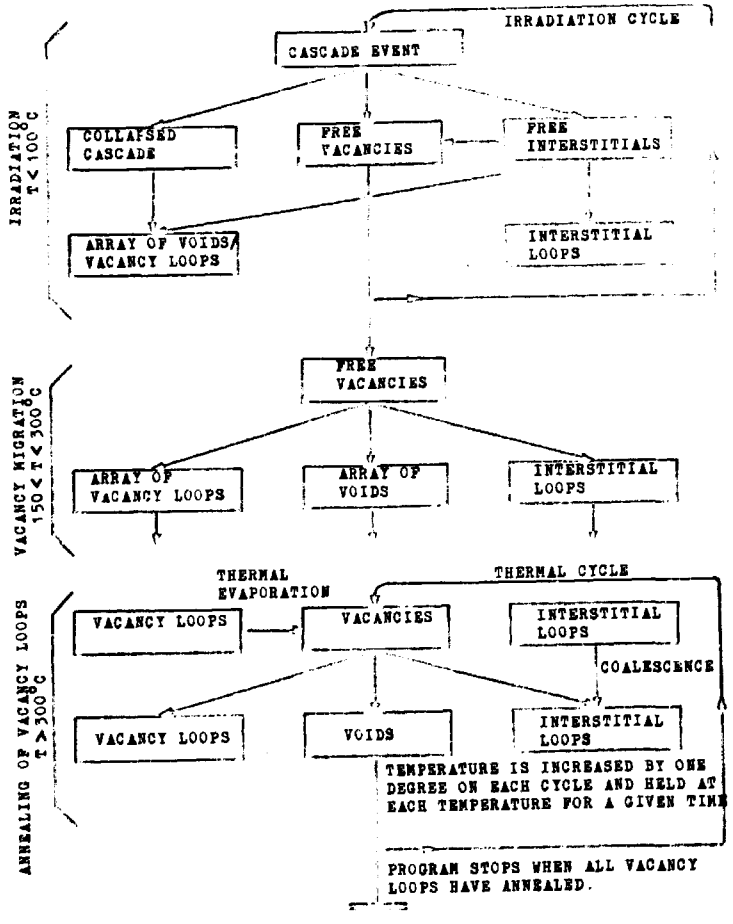


Figure 1.

Simplified flow diagram of the computer program showing the processes occuring during irradiation and subsequent annealing.

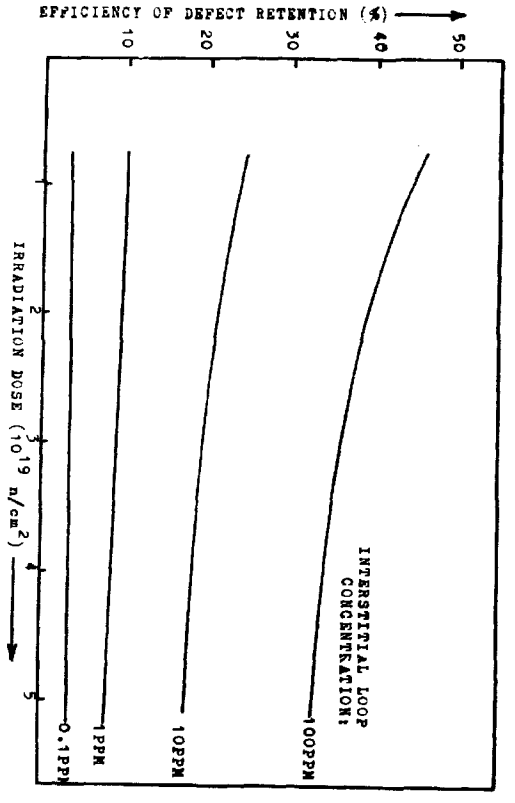


Figure 2.

Variation of efficiency of defect retention with dose as function of interstitial loop concentration.

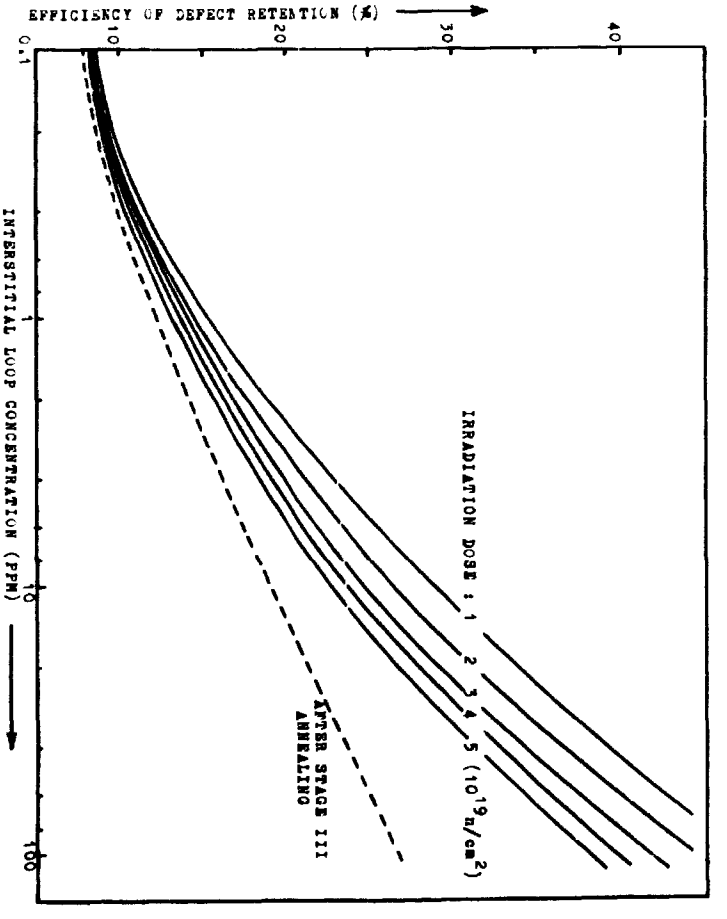


Figure 3.

Variation of efficiency of defect retention with interstitial loop concentration as function of irradiation dose. Dotted line shows efficiency for dose of $5.10^{19}$ n/cm$^2$ after stage 3 vacancy annealing.

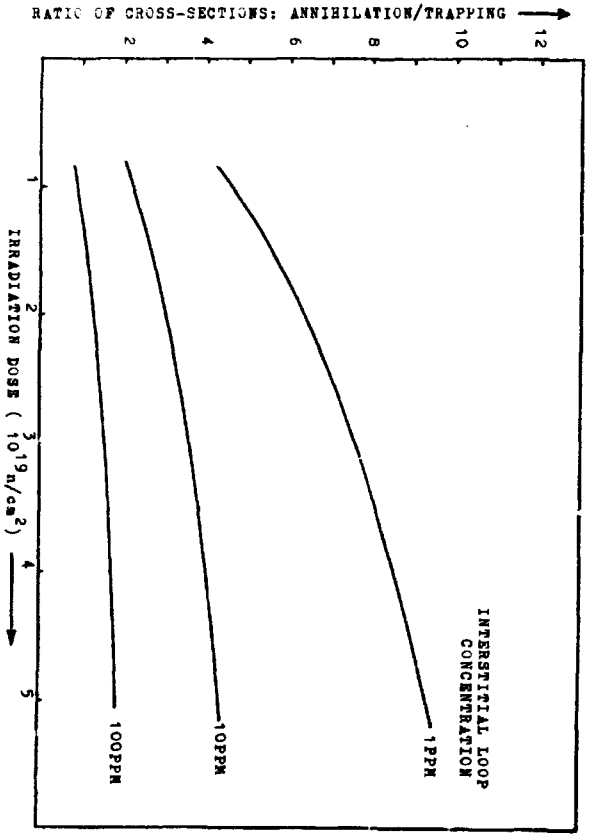


Figure 4.

Ratio of annihilation to trapping cross-section of interstitials migrating during irradiation as function of irradiation dose and interstitial loop concentration.

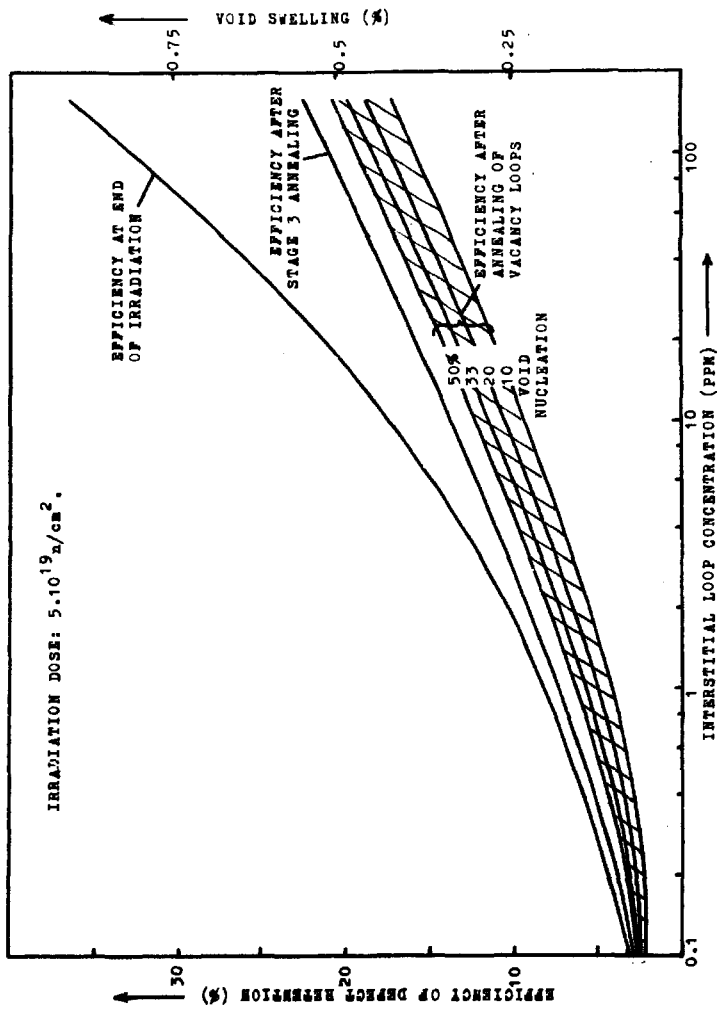


Figure 5. Efficiency of defect retention (and void swelling) after irradiation of $5.10^{19}$ n/cm$^2$, plus high temperature annealing, as function of void nucleation in cascade and original interstitial loop nucleation.

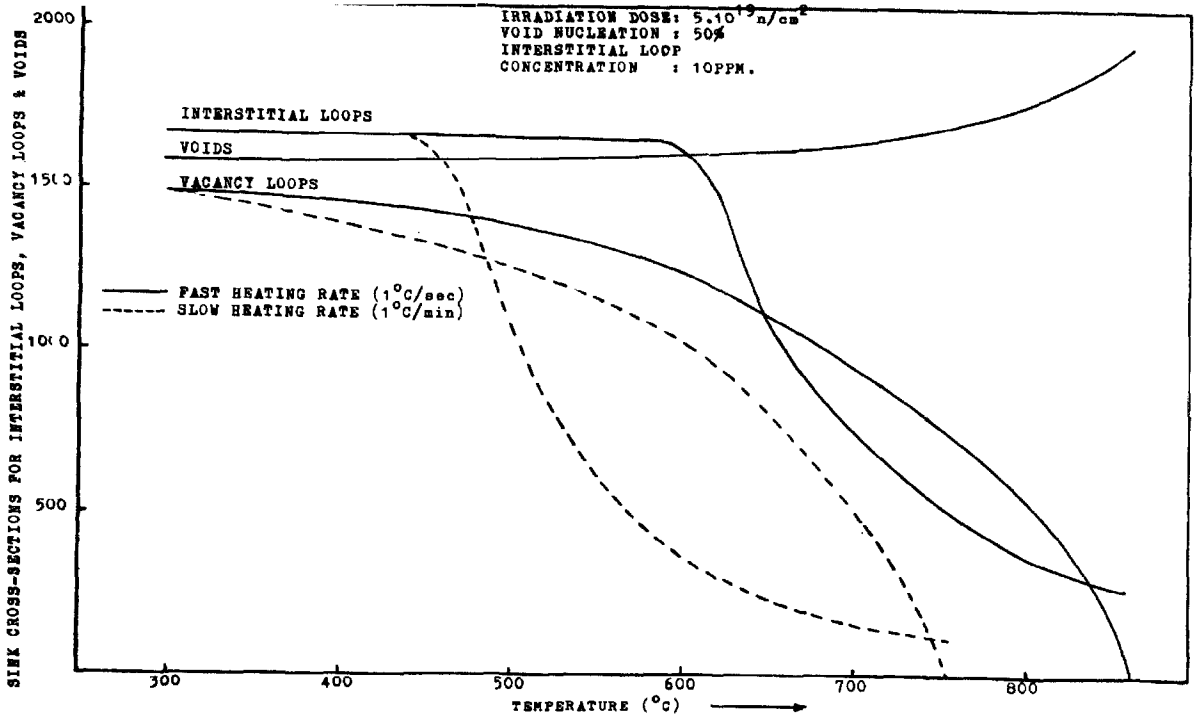


Figure 6.

Variations of sink cross-sections (Å per million atoms) for interstitial loops, vacancy loops and voids during post-irradiation annealing.

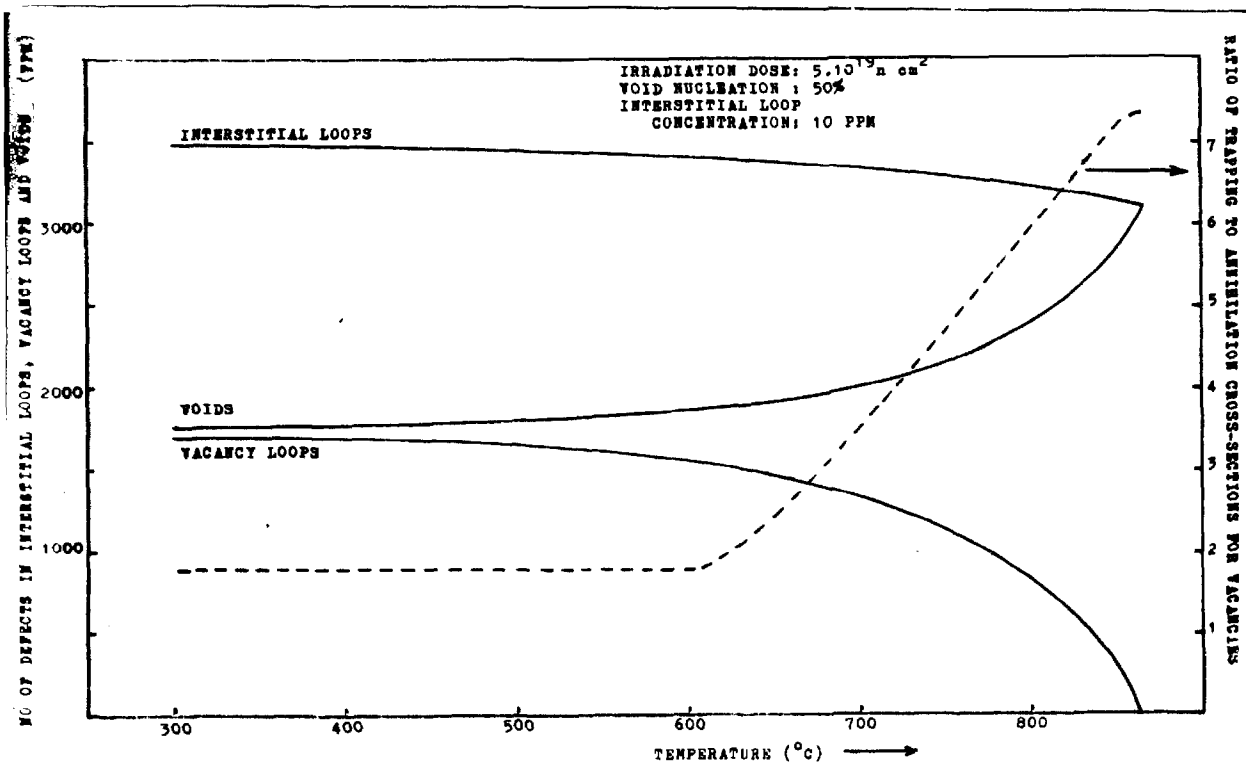


Figure 7.

Variations of defect numbers in interstitial loops, vacancy loops and void during annealing. Also plotted is the ratio of trapping to annihilation cross-section for vacancies.