**MODELING OF POINT DEFECT CLUSTER EVOLUTION UNDER PULSED IRRADIATION**

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## OBJECTIVE

Most of the theoretical investigations of damage accumulation in irradiated materials have focused on steady irradiation with little attention paid to pulsed irradiation conditions. However, the problem of pulsed irradiation is quite important when one considers that pulsing is an inherent aspect of current and proposed irradiation facilities such as the MTS (LANL), SNS (ORNL), IFMIF, and ITER. Some theoretical work along this line that has been done in the past predicted essential differences between pulsed and steady irradiations. However exact calculations of damage accumulation under pulsed irradiation have never been done. This work intends to fill this gap.

## SUMMARY

The main results of this work are summarized as follows:

- For the first time point defect (PD) cluster nucleation and growth during different pulsed irradiation regimes has been calculated and compared with that of steady irradiation.
- It is found that PD cluster nucleation is extremely sensitive to the type of irradiation: it is enhanced in the case of pulsed irradiation. The effect increases with decrease of pulse frequency.
- Rate of damage accumulation under pulsed irradiation is larger at small doses and smaller at high doses than that for steady irradiation.
- The calculation results obtained suggest that the pulse structure of MTS and IFMIF may reproduce damage accumulation quite close to that one may expect for steady irradiation whereas it may be very different in the case of SNS irradiation.
- The new method for numerical integration of the Master Equation used provides a valuable tool to link experimental data obtained using different types of irradiations.

## PROGRESS AND STATUS

### I. Introduction

Nucleation, growth and coarsening of point defect clusters or secondary phase precipitates are responsible for numerous changes that occur in the physical and mechanical properties of materials during irradiation. Thus, it is important to develop a fundamental understanding of material behaviour under pulsed irradiation conditions, and the differences between this and the steady irradiation characteristic of fission reactors in which a substantial amount of irradiation data has been obtained. It is also important to evaluate the influence of pulsed irradiation vis-à-vis steady irradiation to determine the validity of data obtained from pulsed sources.

Computational modelling can provide the required critical evaluation. In the late seventies and early eighties work was done to elucidate the impact of pulsed irradiation on microstructure evolution in comparison to that of steady irradiation [1-4]. It was shown that there is quite a large difference in

microstructure evolution between the two types of irradiation, however the calculations were only done for very low doses. In this work we report the results for modelling of nucleation and growth of voids in the dose range up to 0.1 dpa.

A detailed cluster dynamics model based on the mean field rate theory has been employed to investigate the influence of pulsed irradiation on void evolution. The material response to pulsed and continuous irradiation conditions has been characterized using a new numerical method for solving the kinetic equations describing point defect cluster dynamics.

## II. Model description

In this work we consider a particular case, namely void formation from vacancies in a pure metal under irradiation with the following assumptions.

1. Primary damage is produced in the form of Frenkel pairs, which are single vacancies and self-interstitial atoms (SIAs).
2. The point defects execute a three-dimensional random walk.
3. The nucleation of vacancy clusters (voids) proceeds via a homogeneous mechanism that is due to vacancy + vacancy reactions.
4. The vacancy clusters are immobile.

These assumptions, however, are only used for simplification and do not lead to any restrictions in the applicability of the analysis presented below.

Within the framework described above, we can write the following Master Equation (ME) for the size distribution of voids (SDF), *f(x,t)* where x is the number of vacancies in a void

$$\frac{\partial f(x,t)}{\partial t} = J(x-1,t) - J(x,t) \tag{1}$$

In Eq. (1) *J (x,t)* is the flux of clusters in space of cluster size:

$$J(x,t) = P(x,t)f(x,t) - Q(x+1,t)f(x+1,t), \tag{2}$$

where *P(x,t)* is the rate of vacancy absorption by a cluster of size x and *Q(x,t)* is the sum of corresponding rates of SIA absorption and vacancy emission from the void. These rates depend on the concentration of the cluster and diffusion properties of the mobile defects. For three-dimensional diffusing point defects they take the following form:

$$\begin{gathered} P(x) = wx^{1/3} D_v C_v(t), \\ Q(x) = wx^{1/3}\left[D_i C_i + D_v \exp\left(-E_v^b(x)/kT\right)\right] = Q^i(x) + Q^v(x), \end{gathered} \tag{3}$$

where, $w = (48\pi^2/\Omega^2)^{1/3}$, $C_v(t), C_i(t)$ and $D_v, C_v(t), C_i(t) D_i$ are the concentrations and diffusion coefficients of vacancies (subscript v) and SIAs (subscript i), respectively; $E_v^b(x)$ is the binding energy of a vacancy with a void of size x, $k_B$ is the Boltzmann constant, T is the absolute temperature and Ω is the atomic volume.

In order to complete the system of equations describing void evolution, we must add equations for the evolution of the point-defect concentrations, $C_v$ and $C_i$, which are given by (see e.g. [5])

$$\begin{aligned}\frac{dC_v(t)}{dt} &= G(t) - \mu_R D_i C_i(t) C_v(t) - D_v(C_v - C_{v0}) Z_v k_v^2 - 2P(1) f(1,t) \\ &\quad + 2Q_v(2) f(2,t) + Q_i(2) f(2,t) \\ &\quad - \sum_{x=1}^{x=\infty} \left( P(x) f(x,t) - Q_v(x+1) f(x+1,t) \right), \\ \frac{dC_i(t)}{dt} &= G(t) - \mu_R D_i C_i(t) C_v(t) - D_i C_i Z_i k_i^2 - \sum_{x=1}^{x=\infty} Q_i(x) f(x,t),\end{aligned} \tag{4}$$

where G(t) is the point defect generation rate, $\mu_R$ is the recombination constant, and $k_v^2$ and $k_i^2$ are the sink strengths of crystal defects (excluding that of voids) for vacancies and SIAs, respectively. Note that the terms containing *P(x)* and *Q(x)* in Eqs. (4) are consistent with those in Eqs. (2) and (3).

The initial and boundary conditions are taken to have the following form:

$$\begin{aligned} & f(x, t=0) = C_{v0}\delta(x-1), \\ & C_v(t=0) = C_{v0}, \\ & C_i(t=0) = 0, \\ & \\ & f(x=1,t) = C_v(t), \\ & f(x=\infty,t) = 0. \end{aligned} \tag{5}$$

where $C_{v0}$ is the thermal equilibrium vacancy concentration. Eqs. (1) - (4) are a set of coupled nonlinear equations and, in the general case, can be solved using numerical methods only.

The main problem in such calculations arises due to the large number of equations to be solved since ME (1) represents a set of equations describing evolution of each size void. To overcome the problem several methods on numerical integration of the above equations have been suggested. A grouping method, first suggested by Kiritani, provided an idea as to how the number of equations may be substantially reduced. A grouping developed by Golubov et al. [6], which provides the most accurate grouping method, is employed here. The method is based on an approximation of the SDF by a linear function within a group of sizes

$$f_i(x) = L_0^i \left( x - \langle x \rangle_i \right) + L_1^i. \tag{6}$$

The clusters are grouped within of a group of widths $\Delta x_i = x_i - x_{i-1}$ which include the clusters of the sizes

$$x = x_{i-1} + k, \ \left( k = 1,..,\Delta x_i \right), \tag{7}$$

where the subscript $i$ indicates the number of a group. Thus each group consists of $n_i = \Delta x_i$ numbers of different sizes. It can be shown that the mean sizes of clusters within an *i* group, $< x >_i$, are equal to

$$< x >_i = x_i - \frac{1}{2}\left( \Delta x_i - 1 \right). \tag{8}$$

Note that in a case when $\Delta x_i = 1$ the mean size is equal to $< x_i >= x_i$. It was shown [5] that the equations for the coefficients $L_0^i$ and $L_1^i$ has the following form

$$\frac{dL_0^i}{dt} = \frac{1}{\Delta x_i}\left[J(x_{i-1}) - J_x(x_i)\right],$$
$$\frac{dL_1^i}{dt} = -\left(\frac{\Delta x_i - 1}{2\sigma_i^2 \Delta x_i}\right)\left\{ J_x(x_{i-1}) + J(x_i) - 2J(< x_i > -\frac{1}{2}) \right\}. \quad (9)$$

where $\sigma_i^2$ is dispersions of cluster sizes in the group, which are given by

$$\sigma_i^2 = \frac{1}{\Delta x_i}\left[\sum_{\alpha=x_{i-1}+1}^{x_i} \alpha^2 - \frac{1}{\Delta x_i}\left(\sum_{\alpha=x_{i-1}+1}^{x_i} \alpha\right)^2\right] \quad (10)$$

Note that the grouping procedure is usually applied for large clusters (i.e. x >>1) when it becomes more practical to replace, with adequate accuracy, the sums in equation (10) by corresponding integrals. In this case, equation (10) is reduced to $\sigma_i^2 = \left(\Delta x_i\right)^2 / 12$.

Eqs. (9) describe the evolution of the SDF with the group approximation. These equations satisfy the conservation laws for the number of voids and the number of vacancies in voids. Note that Eqs. (9) are identical to equation (1) in the limiting case when $\Delta x_i = 1$. Because of this there is no problem in describing the SDF in the region between the ungrouped and grouped clusters.

**III. Pulse structure and irradiation and material parameters.**

Pulsed irradiation used for the analysis is taken in a form schematically presented in Fig. 1.

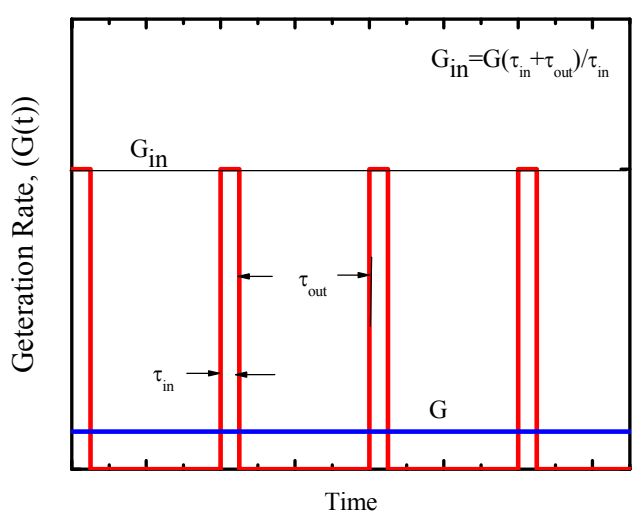


Fig. 1. Pulsed irradiation schematic. $\tau_{in}$ and $\tau_{out}$ are the duration of a pulse and time between the pulses; $G_{in}$ and $G$ are the rate of point defect generation in the pulse regime and that in the equivalent steady irradiation.

Essential parameters of a pulsed irradiation are the duration of pulses, $\tau_{in}$, and the frequency, $\omega$, which is determined as follows

$$\omega = \left( \frac{1}{\tau_{in} + \tau_{out}} \right)_{|\tau_{out}/\tau_{in} >> 1} \approx \frac{1}{\tau_{out}}. \qquad (11)$$

In this work the calculations have been done for steady irradiation and two pulsed irradiations with $\tau_{in}$=7.5x10$^{-4}$ s and the frequency given in Table 2. Material and irradiation parameters are presented in Table 2.

Table 1. Pulsed Irradiation characterization.

| $\tau_{out}/\tau_{in}$ | ω (Hz) |
|---|---|
| 100 | 13.20 |
| 1000 | 0.132 |

Table 2. Material and irradiation parameters used in calculations.

| | |
|---|---|
| Atomic volume, Ω | $1.189 \times 10^{-29}$ m$^3$ |
| Recombination coefficient | $1.0 \times 10^{21}$ m$^{-2}$ |
| Vacancy diffusion coefficient:<br>Preexponent,<br>Migration energy, | <br>$5.0 \times 10^{-5}$ m$^2$/s<br>0.90 eV |
| SIA diffusion coefficient:<br>Preexponent,<br>Migration energy, | <br>$1.0 \times 10^{-6}$ m$^2$/s<br>0.25 eV |
| Capture efficiencies of dislocations for vacancies and SIAs | 1.0, 1.04 |
| Dislocation density, $\rho_d$ | $1\times 10^{14}$ m$^{-2}$ |
| Temperature, T | 473 to 673 K |
| Displacement rate, *G* | $1.0\times 10^{-6}$ dpa/s |
| Terminal irradiation dose | 0.1 dpa |

## IV. Results of Calculations

The calculated results for the case of the steady irradiations are shown in Fig. 2 - 4. As can be seen from Figs. 2 swelling obeys a typical behaviour for the case of homogeneous nucleation: the density of voids is very high at low temperature and goes down very fast with temperature increase. Consequently, temperature dependence of swelling (see inset plot in Fig. 2a) shows a maximum at temperature about 623 K and rapidly decreases with temperature increase.

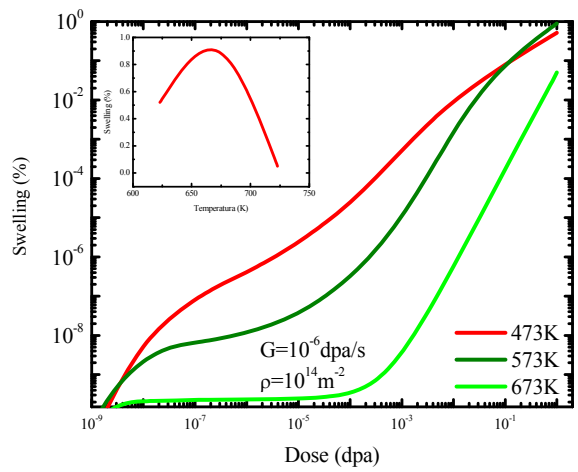


Fig. 2a. Dose dependence of swelling at different temperatures.

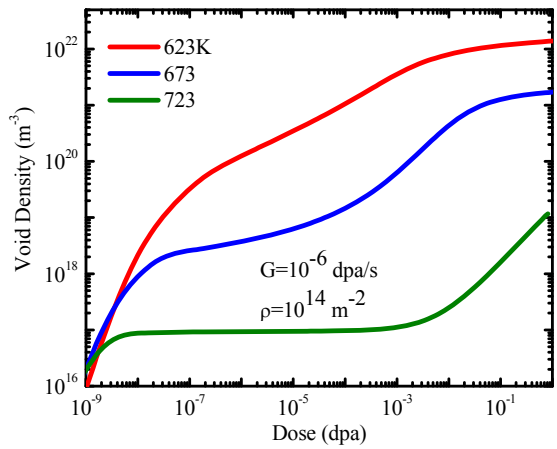


Fig. 2b. Dose dependence of void density at different temperatures.

The dose dependence of the point defect concentrations and mean diameter of voids are presented in Figs. 3. The SDFs of voids at the terminal dose of 0.1 dpa are presented in Fig. 3. As can be seen from Fig. 3a, PD concentrations decrease with dose increases after reaching maximums at temperatures 623 and 673 K. In contrast, they do not practically depend on the irradiation dose at T=723 K because the sink strength of voids at all doses is much smaller that that of dislocations (see Fig. 5). Note that a shape of SDF at this temperature obeys a typical behaviour for a regime when continuous void nucleation takes place at constant nucleation rate (see [6]).

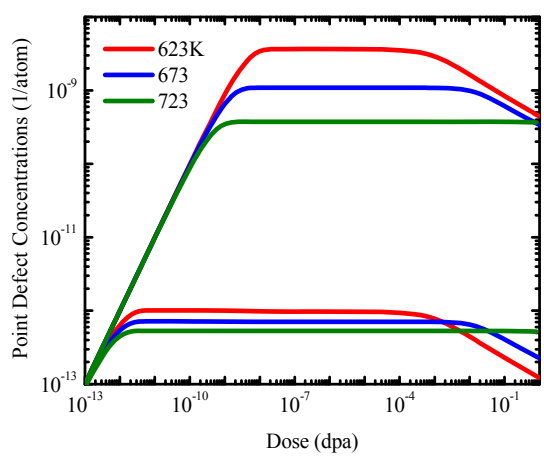


Fig. 3a. Dose dependence of point defect concentrations at different irradiation temperatures.

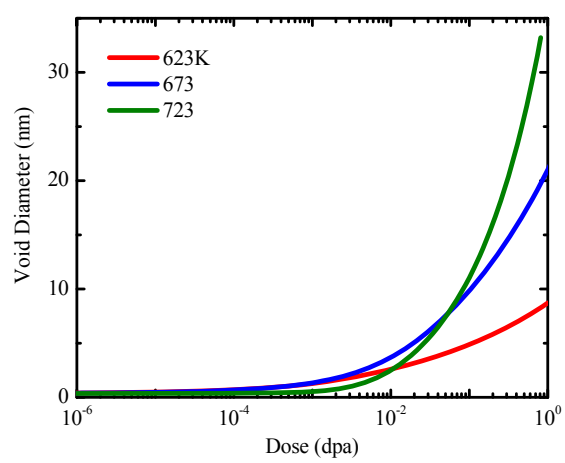


Fig. 3b. Dose dependence of mean diameter of voids at different irradiation temperatures.

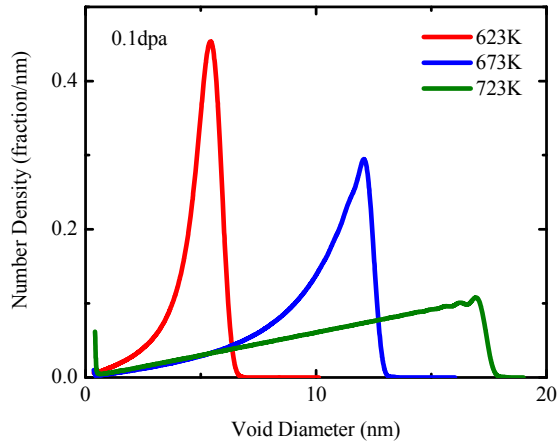


Fig. 4. Size distribution functions at the terminal dose of 0.1 dpa.

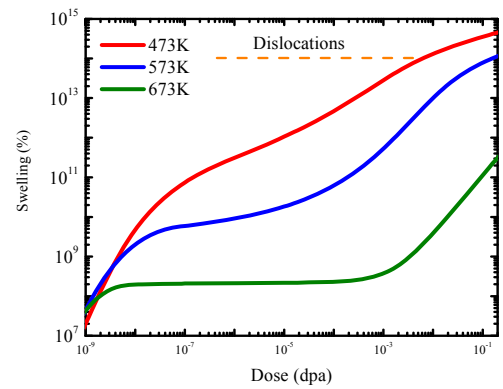


Fig. 5. Dose dependence of sink strengths of voids at different temperatures.

*Pulsed irradiation*

The calculations in this case are more time consuming in comparison with that of the steady irradiation since the time step for numerical integration, $\Delta t$, of the ME has to be extremely small. Indeed, it is quite obvious that the magnitude of $\Delta t$ has to be smaller than the pulse duration, $\Delta t < \tau_{in}$, otherwise the pulse irradiation structure would be lost. Our calculations show that in fact it has to be much smaller, i.e. smaller by at least one order of magnitude. Moreover it is very sensitive to the shape of a pulse. The pulse shape presented in Fig. 1 requires the time step to be smaller than $\tau_{in}$ by about two orders of magnitudes. The time step may be larger in the case when the pulse shape is not so abrupt. In our calculations the pulse shape is taken in the form of a Gaussian

$$G(t) \sim \frac{G_{in}}{\sqrt{2\pi}\tau_{in}} \exp\left(-\left(\frac{t - n\tau_{out}}{\tau_{in}}\right)^2\right), \qquad n\tau_{out} - \alpha\tau_{in} \le t \le n\tau_{out} + \alpha\tau_{in}, \tag{12}$$

where the integer number $n$ is the pulse number; a parameter α is a number equal to 2 to 3 used to make the pulse form smoother. It has been found that for pulse duration of $7.5\text{x}10^{-4}$ second an appropriate time step may be chosen to be equal to$10^{-5}$ second. Indeed, as can be seen in Fig. 6 further decrease of the time step does not practically affect the void SDF.

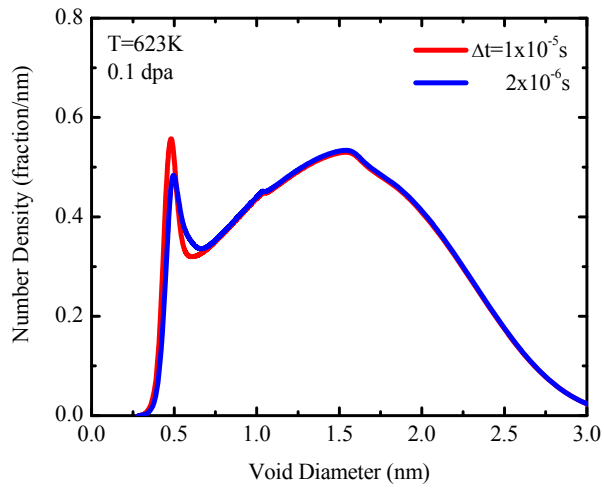


Fig. 6. Terminal size distribution function of voids calculated using two different time steps, $10^{-5}$ and $2x10^{-6}$ second.

Because the calculations in the case of pulsed irradiation are very time consuming they have been carried out for the case corresponding the maximum in swelling, i.e. for irradiation temperature equal to 673 K, only. The dose dependence of PD concentration calculated for the cases of ω=13.2 and 0.132 Hz are presented on Figs. 7. As can be seen from the plots the variation of PD concentration with the irradiation dose is quite different compared to the steady irradiation.

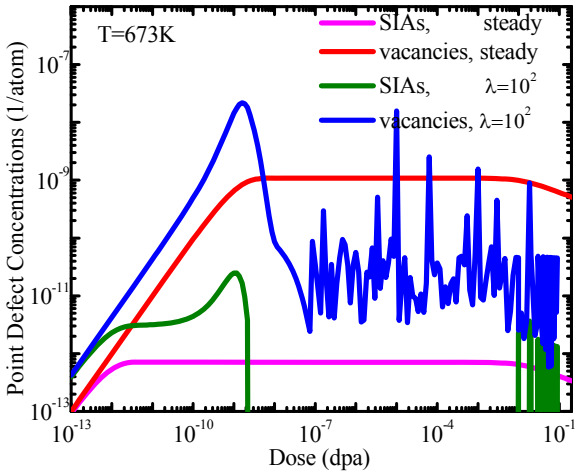


Fig. 7a. Dose dependence of PD concentration calculated for ω=13.2 Hz.

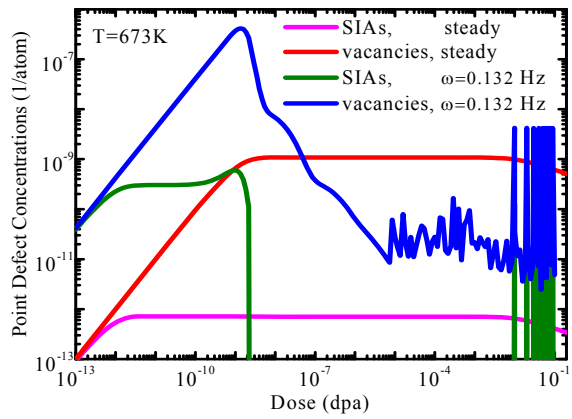


Fig. 7b. Dose dependence of PD concentration calculated for ω=0.132 Hz.

As can be seen the vacancy and SIA concentrations show very large fluctuation with dose and are much higher than those for the corresponding steady irradiation. Note that the SIA concentration is practically zero most the time due to their fast diffusion.

The dose dependence of the void number density and the number of vacancies accumulated in voids are presented in Figs. 8. In accordance with the much higher vacancy concentration in the case of the pulsed irradiation, the void density in this case is much higher as well. It leads to a much higher level swelling at relatively small doses. However this tendency is reversed at higher doses: the rate of swelling

accumulation under pulsed irradiation becomes smaller than that of steady irradiation with the effect much more pronounce in the case the low frequency, ω=0.132 Hz. The reason may be understood from a comparison of the SDFs for those three cases presented in Fig. 9. Indeed as can be seen from Fig. 9a the striking difference between the case of ω=0.132 Hz and other cases is an appearance of an extra peak at small sizes (see more detailed shape of the SDFs in Fig. 9b). It is probably the reason for swelling decrease under pulse irradiation because this group of small voids, which are unstable, competes with the stable ones. Some support for this explanation is given below.

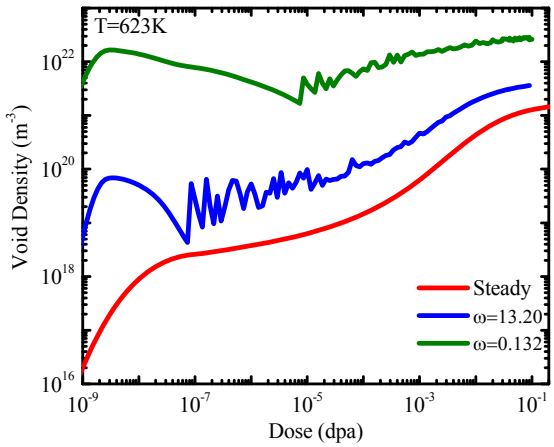


Fig. 8a. Dose dependence of void density calculated for ω=13.2 Hz.

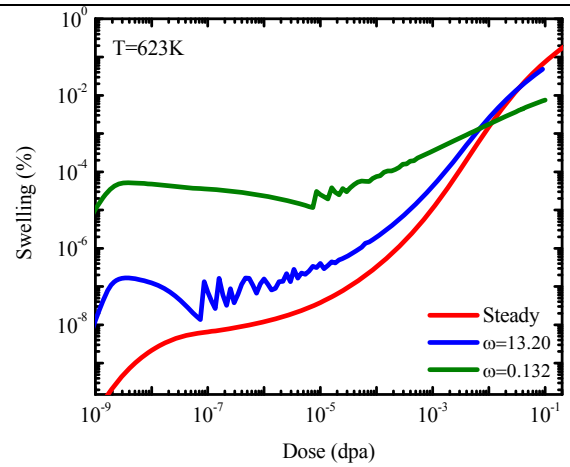


Fig. 8b. Dose dependence of swelling calculated for ω=0.132 Hz.

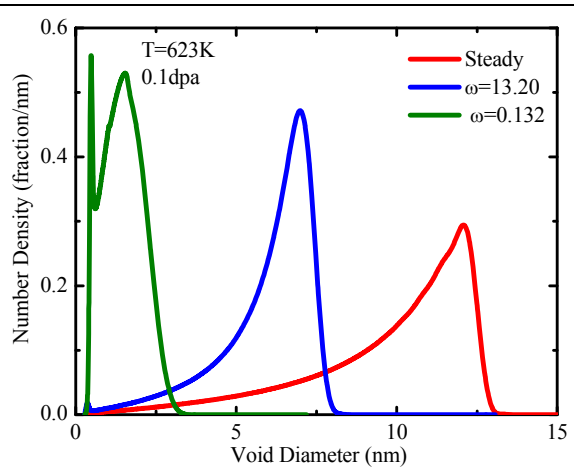


Fig. 9a. Terminal size distribution functions of voids calculated for steady and pulse irradiations.

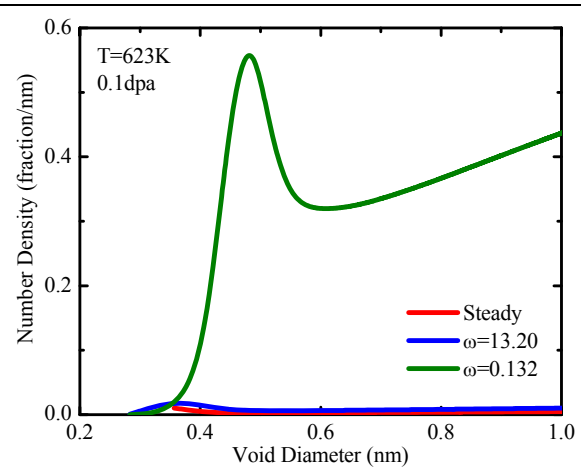


Fig. 9b. The same as Fig.9 in the range of sizes up to 1 nm.

In order to elucidate the point an extra calculation has been done for the case ω=0.132 Hz when the recombination constant is taken to be zero. This case has been considered with an aim to exclude the mutual recombination between vacancies and SIAs that helps to narrow down the number of mechanisms responsible for the effect. The dose dependence of PD concentrations and swelling for the

case together with that when the recombination is taken into account are presented in Figs. 10. As can be seen from Fig. 10a the PD concentrations are higher in the case of "no recombination". The magnitude of swelling at low doses is also higher than that when recombination is taken into account. However the swelling rate at the higher doses is strongly decreased. Comparison of SDFs for those two cases presented in Fig. 11 shows that the fraction of small unstable voids in the case of "no recombination" is higher than that for the case with recombination. Because in the case of "no recombination" the only mechanism involved in the void evolution is a partitioning of point defects between voids and dislocations, one may conclude that the unstable small voids are really responsible for the swelling decrease under the pulsed irradiation.

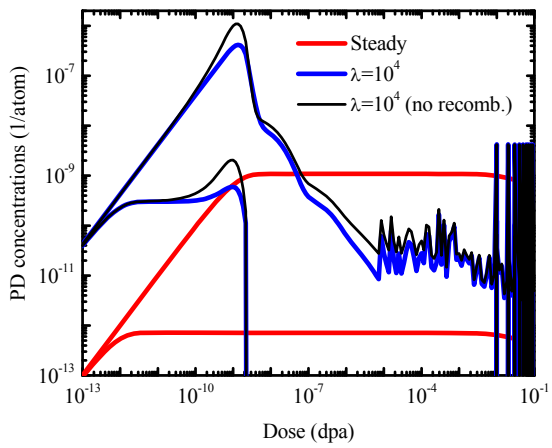


Fig. 10a. Dose dependence of PD concentration calculated with and without recombination for ω=0.132 Hz.

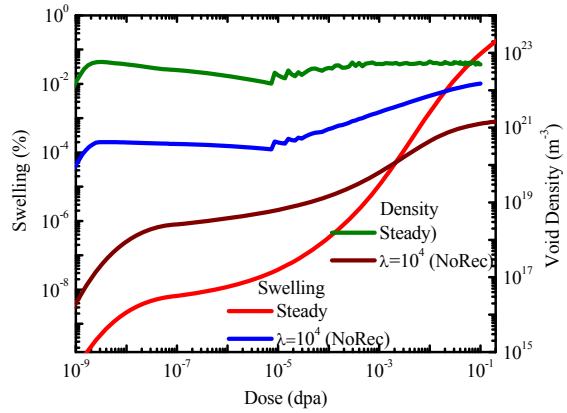


Fig. 10b. Dose dependence of swelling calculated with and without recombination for ω=0.132 Hz.

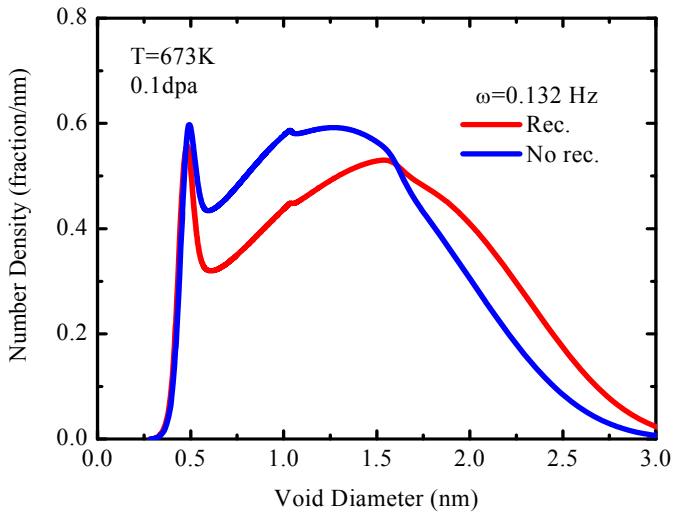


Fig. 11. Terminal size distribution functions of voids calculated for the pulsed irradiation calculated with and without recombination (ω=0.132 Hz).

The same conclusion results from comparing the dose dependence of void swelling calculated in the case of steady irradiation with that which can be calculated analytically for the case of the pulsed irradiation. Indeed it is very well established that in the case without recombination the swelling rate is given by

$$\frac{dS}{d(Gt)} = \frac{Z_i - Z_v}{Z_v} \frac{k_v^2 \rho}{\left(k_v^2 + \rho\right)^2}, \tag{13}$$

where $k_v^2$ is the sink strength of voids. Since the dose dependence of $k_v^2$ in the case "no recombination" is calculated, the swelling rate given by Eq. (13) can be compared with those calculated for the cases of steady irradiation and "no recombination". Such a comparison is shown in Fig. 12. As can be seen from the plot Eq. (13) does not provide a correct description of the case under consideration.

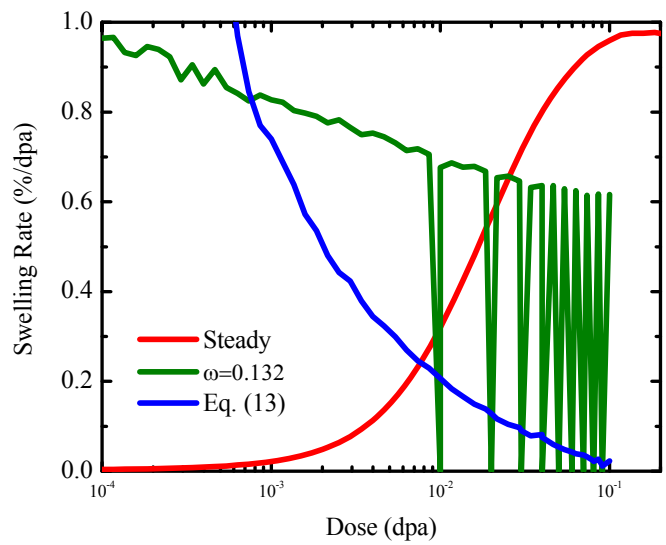


Fig. 12. Dose dependence of swelling rate calculated with and without recombination for ω=0.132 Hz.

## V. Conclusions

The calculations presented in this work revealed that the damage accumulation in the form of voids under pulsed irradiation may be qualitatively different compared to that taking place under steady irradiation with the same average rate of PD generation. It is shown that in the case considered here the void density is orders of magnitude higher that that of the steady irradiation and the swelling rate is dramatically decreased at high enough doses. Moreover, it is shown that the damage accumulation cannot be explained by using the ordinary mean size approximation widely used for the case of steady irradiation. In the other words, the numerical integration of ME is the only method which can provide an appropriate description of the damage accumulation under pulsed irradiation.

## ACKNOWLEDGEMENTS

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