Effect of pulse irradiation on the evolution of damage structure

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Abstract

Charged particle beams from accelerators driven by radiofrequency or with beam scanners possess time structures (i.e., pulse beam operations). However, it is not clear whether the irradiation effects of these pulse beam operations are the same as those of continuous beams, although the average damage rate is the same in both cases. In this study, the difference between continuous beams and pulse beams, and the effect of the repetition frequency and pulse duration on the defect accumulation are evaluated by the rate equation analysis for Al and Fe. The growth rate of interstitial type dislocation loops in Al is simulated with fixed duty ratios (constant pulse duration over the irradiation period) at 453 K. The effect of pulse duration on a constant period of 1 s is simulated in Fe, changing the pulse duration from 1 to 10^{-6} s, while the damage (dpa) caused by one pulse remains the same. The results are analyzed and the effects of pulse duration and repetition rate on the damage structure evolution are demonstrated in relation to materials parameters.

Key words

Charged particle irradiation, pulse beam operation, interstitial type dislocation loops, rate equation

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1. Introduction

Irradiation experiments are essential for the development of nuclear materials. High energy charged particles such as ions and electrons, which are produced in accelerators are frequently employed for these experiments. This is because the irradiation conditions and damage rates in the case of ions and electrons can be controlled more easily as compared to those of neutron irradiation, which employs nuclear reactors. Ion/electron beams from accelerators driven by radiofrequency (RF) or accelerators with beam scanners possess time structures (periodic/discontinuous intensities). An example of a pulse train is shown in Fig. 1. The pulse frequency is $1/T$ and H is the beam intensity, but we consider H to be the damage rate (dpa/s) in a pulse duration τ. It is not clear if the irradiation effects of these pulse beam operations are the same as those of continuous beams, although the average damage rate is the same in the two cases. A pulse (macro pulse) consists of many bunches (micro pulses) in the case of RF-driven accelerators, but the effect of these bunches is not considered in our study.

The concentration and migration efficiency of point defects at the initial stage of irradiation are shown schematically in Fig. 2 [1, 2, 3] for intermediate sink density [4]. The migration efficiency is the product of the concentration, C_k, and mobility, M_k, of point defects, k, and it plays an important role in the nucleation and growth of point defect clusters [1]. For example, the growth rate of interstitial type dislocation loops is expressed by $Z_{SI\_I}M_IC_I$ - $Z_{SI\_V}M_VC_V$, where Z, M_IC_I and M_VC_V are the number of reaction sites, the migration efficiencies of interstitials and vacancies, respectively. At the initial stage of irradiation, the concentrations of interstitials and vacancies are the same, but the migration efficiency of interstitials exceeds that of the vacancies by time L_2 in Fig. 2, because of the high mobility of the former. Hence, if the pulse duration L is shorter than time L_2, the pulse train will fulfill the conditions for the interstitial migration efficiency to be dominant. The interstitial dominant conditions during irradiation were demonstrated experimentally by Arai *et al*. [5]. They utilized the scanning ability of high voltage electron microscopy for the generation and observation of irradiation induced defect clusters. They employed two methods. the flip-flop method in which the electron beam reverts to the observation area of the transmission electron microscope intermittently, and the cycle beam method, where the electron beam traces circular and intermittent paths around the observation area.

The point defect process is, however, far more complex, and the accumulation of vacancies and the thermal decomposition of cluster change the defect structure evolution with increasing irradiation. In this paper, the difference between continuous and pulse beams, and the effect of repetition frequency and pulse duration on the defect accumulation are presented. The rate equation analysis is used for evaluating the time structure effect.

2. Method

The model used for the calculations is almost the same as that used in previous studies [6, 7, 8, 9], and is based on the rate theory. It describes the reaction rates among point defects and their defect clusters. The main difference between the present model and the previous ones is that there is no introduction of solute atoms, and only pure metal cases, Al and Fe, are treated.

The following assumptions are made in the calculations:

(1) Mobile defects are interstitials, di-interstitials, vacancies and di-vacancies.

(2) Quadri-interstitials and quadri-vacancies are set for stable nuclei belonging to loops and voids, respectively. The choice of quadri-point defects is for convenience to reduce the number of equations, and to introduce thermal dissociation in di-interstitials, di-vacancies, tri-interstitials and tri-vacancies.

(3) The effect of cascade damage is introduced in the direct formation of di-interstitials and di-vacancies. It is not included in the simulation of electron irradiation as demonstrated in Sections 3.1.

The time dependence of 10 variables is calculated for the following quantities: the concentration of interstitials, C_I, di-interstitials, C_{I2}, tri-interstitials, C_{I3}, interstitial clusters (interstitial-type dislocation loops), C_{IC}, vacancies, C_V, di-vacancies, C_{V2}, tri-vacancies, C_{V3}, vacancy clusters (voids), C_{VC}, total interstitials in interstitial type dislocation loops, R_{IC}, and total vacancies in voids, R_{VC}. The rate equations are expressed as follows:

$$\frac{dC_I}{dt} = P(t) + 2Z_{I2B}B_{I2}M_IC_{I2} + Z_{I3B}B_{I3}M_IC_{I3} - 2Z_{I\_I}C_IM_IC_I - Z_{V\_I}C_VM_IC_I$$
$$- Z_{I2\_I}C_{I2}M_IC_I - Z_{2V\_I}C_{V2}M_IC_I - Z_{I3\_I}C_{I3}M_IC_I - Z_{V3\_I}C_{V3}M_IC_I - Z_{SI\_I}S_IM_IC_I$$
$$- Z_{SV\_I}S_VM_IC_I - C_SM_IC_I + Z_{V\_I2}C_VM_{I2}C_{I2} + Z_{I3\_2V}C_{I3}M_{V2}C_{V2} ,$$

$$\frac{dC_V}{dt} = P(t) + 2Z_{V2B}B_{V2}M_VC_{V2} + Z_{V3B}B_{V3}M_VC_{V3} + Z_{V2\_I}C_{V2}M_IC_I - Z_{V\_I}C_VM_IC_I - 2Z_{V\_V}C_VM_VC_V$$
$$- Z_{V\_I2}C_VM_{I2}C_{I2} - Z_{V2\_V}C_{V2}M_VC_V - Z_{I3\_V}C_{I3}M_VC_V - Z_{V3\_V}C_{V3}M_VC_V$$
$$- Z_{SI\_V}S_IM_VC_V - Z_{SV\_V}S_VM_VC_V - C_SM_VC_V + Z_{V3\_I2}C_{V3}M_{I2}C_{I2} ,$$

$$\frac{dC_{I2}}{dt} = A_{IC}P(t) - Z_{I2B}B_{I2}M_IC_{I2} + Z_{I3B}B_{I3}M_IC_{I3} + Z_{I\_I}C_IM_IC_I - Z_{I2\_I}C_{I2}M_IC_I + Z_{I3\_V}C_{I3}M_VC_V$$
$$- Z_{V\_I2}C_VM_{I2}C_{I2} - 2Z_{I2\_I2}C_{I2}M_{I2}C_{I2} - Z_{V2\_I2}C_{V2}M_{I2}C_{I2} - Z_{I3\_I2}C_{I3}M_{I2}C_{I2}$$
$$- Z_{V3\_I2}C_{V3}M_{I2}C_{I2} - Z_{SI\_I2}S_IM_{I2}C_{I2} - Z_{SV\_I2}S_VM_{I2}C_{I2} - C_SM_{I2}C_{I2} ,$$

$$\frac{dC_{V2}}{dt} = A_{VC}P(t) - Z_{V2B}B_{V2}M_VC_{V2} + Z_{V3B}B_{V3}M_VC_{V3} - Z_{V2\_I}C_{V2}M_IC_I + Z_{V3\_I}C_{V3}M_IC_I$$
$$+ Z_{V\_V}C_VM_VC_V - Z_{V2\_V}C_{V2}M_VC_V - Z_{V2\_I2}C_{V2}M_{I2}C_{I2} - 2Z_{V2\_V2}C_{V2}M_{V2}C_{V2}$$
$$- Z_{I3\_V2}C_{I3}M_{V2}C_{V2} - Z_{V3\_V2}C_{V3}M_{V2}C_{V2} - Z_{SI\_V2}S_IM_{V2}C_{V2} - Z_{SV\_V2}S_VM_{V2}C_{V2}$$
$$- C_SM_{V2}C_{V2} ,$$

$$\frac{dC_{I3}}{dt} = -Z_{I3B}B_{I3}M_IC_{I3} + Z_{I2\_I}C_{I2}M_IC_I - Z_{I3\_I}C_{I3}M_IC_I - Z_{I3\_V}C_{I3}M_VC_V - Z_{I3\_I2}C_{I3}M_{I2}C_{I2} - Z_{I3\_V2}C_{I3}C_{V2}M_{V2} ,$$

$$\frac{dC_{V3}}{dt} = -Z_{V3B}B_{V3}M_VC_{V3} - Z_{V3\_I}C_{V3}M_IC_I + Z_{V2\_V}C_{V2}M_VC_V - Z_{V3\_V}C_{V3}M_VC_V - Z_{V3\_I2}C_{V3}M_{I2}C_{I2} - Z_{V3\_2V}C_{V3}M_{V2}C_{V2} ,$$

$$\frac{dC_{IC}}{dt} = Z_{I3\_I}C_{I3}M_IC_I + Z_{I2\_I2}C_{I2}M_{I2}C_{I2} + Z_{I3\_I2}C_{I3}M_{I2}C_{I2} ,$$

$$\frac{dC_{VC}}{dt} = Z_{V3\_V}C_{V3}M_VC_V + Z_{V2\_V2}C_{V2}M_{V2}C_{V2} + Z_{V3\_V2}C_{V3}M_{V2}C_{V2} ,$$

$$\frac{dR_{IC}}{dt} = 4Z_{I3\_I}C_{I3}M_IC_I + Z_{SI\_I}S_IM_IC_I - Z_{SI\_V}S_IM_VC_V + +4Z_{I2\_I2}C_{I2}M_{I2}C_{I2} + 5Z_{I3\_I2}C_{I3}M_{I2}C_{I2} + 2Z_{SI\_I2}S_IM_{I2}C_{I2} - 2Z_{SI\_V2}S_IM_{V2}C_{V2} ,$$

$$\frac{dR_{VC}}{dt} = -Z_{SV\_I}S_VM_IC_I + 4Z_{V3\_V}C_{V3}M_VC_V + Z_{SV\_V}S_VM_VC_V - 2Z_{SV\_I2}S_VM_{I2}C_{I2} + 4Z_{V2\_V2}C_{V2}M_{V2}C_{V2} + 5Z_{V3\_V2}C_{V3}M_{V2}C_{V2} + 2Z_{SV\_V2}S_VM_{V2}C_{V2} ,$$

$$S_I = 2(\pi R_{IC}C_{IC})^{1/2},$$

$$S_V = (48\pi^2 R_{VC}C_{VC}^2)^{1/3},$$

where P(t) = $H\times F(t)$ is the time-dependent damage rate with a given periodicity. H is the damage rate in a pulse duration (see Fig. 1), and $F(t)$ is a periodic function approximated as a Fourier series with a single frequency over time t. Z is the number of sites in the spontaneous reaction for each process indicated by the corresponding subscript. S is the total sink efficiency of the clusters [1, 3, 10]. The mobility of the defects, M, is expressed as ν exp($-\frac{E_M}{kT}$), where ν, the effective frequency associated with the vibration of the defects in the direction of the saddle point, is taken as 10^{13}/s. In the case of the reaction of two mobile defects, only the defect with higher mobility is considered. For example, M_I+M_V is M_I. E_M, k, and T are the migration energy, the Boltzmann constant and absolute temperature, respectively. AP(t) is the direct formation rate of clusters in cascade processes. B is the dissociation probability of vacancies and interstitials, and it is expressed as ν exp($-\frac{E_B}{kT}$), where E_B is the binding energy. The subscripts I, V, IC and VC denote interstitials, vacancies, interstitial clusters (interstitial type dislocation loops) and vacancy clusters (voids), respectively. Surfaces, grain boundaries, and pre-existing defects such as dislocations are expressed by the sink efficiency, C_S [1, 3, 10]. The concentrations are expressed in fractional units. These terms are almost the same as those in our previous papers [6, 7, 8, 9]. The values of the coefficients used are listed in Table 1 (Fe) and Table 2 (Al). The equations are solved with a SUite of Nonlinear and DIfferential/ALgebraic equation Solvers (SUNDIALS) [11].

3. Results

3.1 Constant τ/T

The growth rate of interstitial type dislocation loops defined as $Z_{SI\_I}M_IC_I + Z_{SI\_I2}M_{I2}C_{I2} - Z_{SI\_V}M_VC_V - Z_{SI\_V2}M_{V2}C_{V2}$ in Al and Fe was simulated with fixed duty ratios (pulse duration over irradiation period: τ/T in Fig. 1.) Fig. 3 illustrates the case of Al irradiated at 453 K with a damage rate in pulse duration, H, of 0.1dpa/s and τ/T = 1/6. These conditions were the same as those in the electron irradiation experiment conducted by Arai *et al*. [5]. Increasing the pulse frequency resulted in an increase in the loop growth rate. The growth rate at low frequency was close to 0.97, as shown by arrow A for the continuous beam irradiation of 0.0167 dpa/s (0.1dpa ×1/6), and that at high frequency was close to 1.9 for continuous beam irradiation of 0.1 dpa/s as shown by arrow B. The open circles in the figure denote the data collected by Arai *et al*. [5]. The absolute values are not compared directly because they depend on Z_{SI_I}, Z_{SI_I2}, Z_{SI_V} and Z_{SI_V2}, but the frequency dependence of the growth rate is simulated well. The loop growth rate saturates at 10^3 Hz. The time for the interstitial concentration and the interstitial migration efficiency constant (quasi-equilibrium state, point L_2 in Fig. 2) is about 1×10^{-3} s, and the corresponding frequency is 1×10^3 Hz. Therefore low frequency (duration longer than point L_2) does not significantly affect the growth rate.

Figure 4 shows the total number of interstitials accumulated in the interstitial type dislocation loops in Fe at 573 K with $H = 1\times10^{-6}$ dpa/s and τ/T = 1/10. At low frequencies, the accumulation of interstitials is close to that in the case of continuous beam irradiation of 1×10^{-7} ($1\times10^{-6}\times1/10$) dpa/s. Under these irradiation conditions, the concentration and migration efficiency of the interstitials reach a maximum value close to 10^{-2} s (L_1 in Fig, 2) and the corresponding frequency of 10^2 Hz. Thus, for frequencies higher than 10^2 Hz, the migration efficiency of the interstitials and their accumulation in the loops are low.

3.2 Constant T and $H\times\tau$

The effect of pulse duration on a constant period of 1 s was simulated. The pulse duration was changed from 1 to 10^{-6} s. In this case the damage (dpa) caused by one pulse remained the same, i.e. $H \times \tau = 1\times10^{-6}$ dpa. For example, the pulse duration of 10^{-3} s corresponds to $H = 1\times10^{-3}$ dpa/s. Fig. 5 illustrates the case of Fe at 473 K, 573 K and 673 K irradiation. Since the pulse period was 1 s, the pulse duration of 1 s corresponded to a continuous beam with a damage rate of 1×10^{-6} dpa/s. An increase in the pulse duration, τ, resulted in an increase in the total number of interstitials accumulated in the loops. When τ is short, H is high and many loops are nucleated. These loops act as the mutual annihilation sites for interstitials and vacancies, and prevent the growth of loops.

3.3 Effect of material parameters and damage rate

As shown in the previous sections, the effects of pulse duration and period are not readily measurable, as these are strongly dependent on the materials parameters and dose rate. For example, the migration efficiency of interstitials reaching the maximum at the time L_1 is expressed as $(Z_{V\_I} PM_I)^{1/2}$ for the simple case of intermediate sink density, and the quasi-equilibrium state at time L_2 is $(M_V C_S)^{-1}$ as indicated in Fig. 2 [2, 3, 4]. Therefore these times depend on the damage rate, mobility of point defects and sink efficiency. Notably L_2 does not directly depend on the damage rate, P. Thus, if the damage rate during duration changes as discussed in Section 3.2, the mutual annihilation rate plays an important role for the nucleation of point defect clusters.

4. Concluding remarks

For comparison of the damage structure evolution by neutron, ion and electron irradiation, past studies typically focused on the damage rate [12] and the deposited energy by one particle [13]. Our simulations demonstrated the importance of pulse duration and repetition rate in the damage structure evolution, which are strongly related to the material parameters. The effect of pulse irradiation should be analyzed, taking into consideration the materials parameters and the irradiation conditions.

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Table 1 Coefficients used in the simulation and the corresponding values in Fe. The unit of *E* is eV.

E_{MI}	E_{MV}	E_{MI2}	E_{MV2}	E_{BI2}	E_{BV2}	E_{BI3}	E_{BV3}	A_{IC}	A_{VC}	C_S	Z_{I_I}	Z_{V_I}	Z_{I2_I}	Z_{V2_I}
0.3	1.2	0.3	2.0	2.0	0.2	2.0	0.045	0.01	0.01	1.0×10^{-9}	10	8	10	8

Z_{I3_I}	Z_{V3_I}	Z_{SI_I}	Z_{SV_I}	Z_{V_V}	Z_{I2_V}	Z_{V2_V}	Z_{I3_V}	Z_{V3_V}	Z_{SI_V}	Z_{SV_V}	Z_{I2_I2}	Z_{V2_I2}	Z_{I3_I2}
10	8	10	8	6	8	6	8	6	8	6	10	8	10

Z_{V3_I2}	Z_{SI_I2}	Z_{SV_I2}	Z_{V2_V2}	Z_{I3_V2}	Z_{V3_V2}	Z_{SI_V2}	Z_{SV_V2}
8	10	8	6	8	6	8	6

Table 2 Coefficients used in the simulation and the corresponding values in Al. The unit of *E* is eV. The parameters not indicated are the same as those of Fe.

E_{MI}	E_{MV}	E_{MI2}	E_{MV2}	A_{IC}	A_{VC}	C_S	E_{BI2}	E_{BV2}	E_{BI3}	E_{BV3}
0.115	0.6	0.115	0.55	0	0	2.0×10^{-4}	2	0.5	0.17	0.4

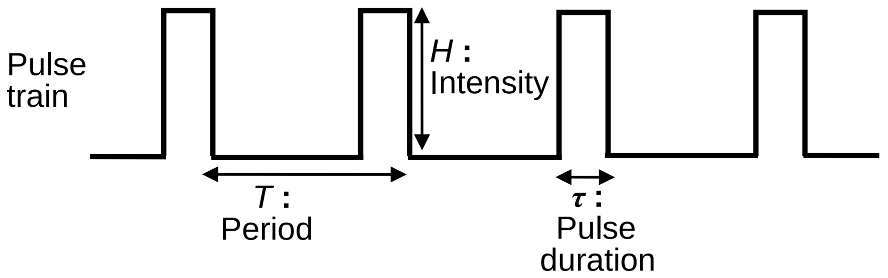

Fig. 1 Example of a pulse train.

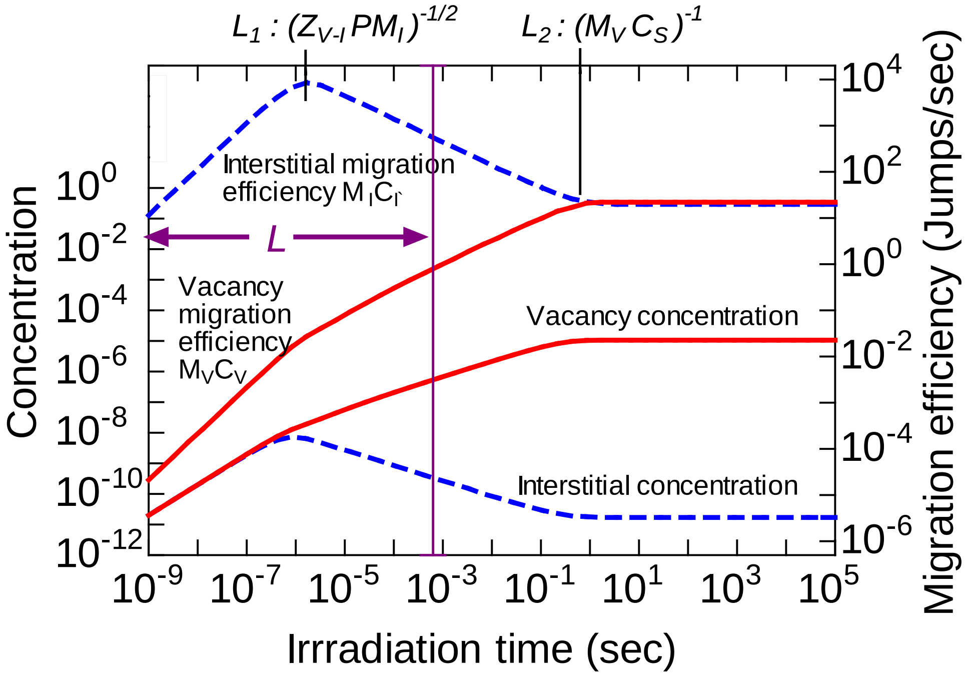

Figure 2 Example of change in the concentration of point defects and their migration efficiencies with irradiation time.

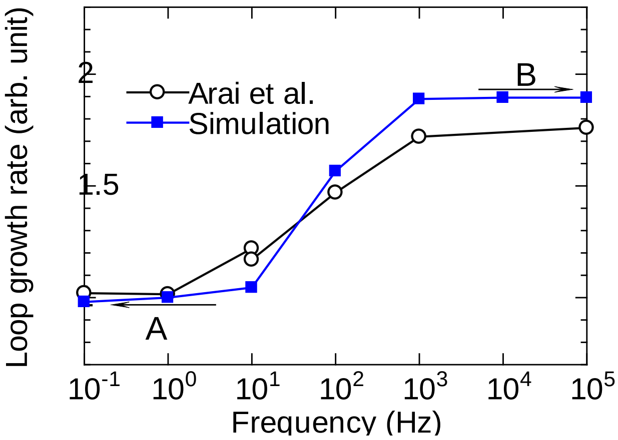

Figure 3 Growth rate of interstitial type dislocation loops in Al. $H = 1\times10^{-1}$dpa/s at 453 K. Solid squares indicate the result of the current study, and circles indicate the result of the experiment carried out by Arai *et al*. [5].

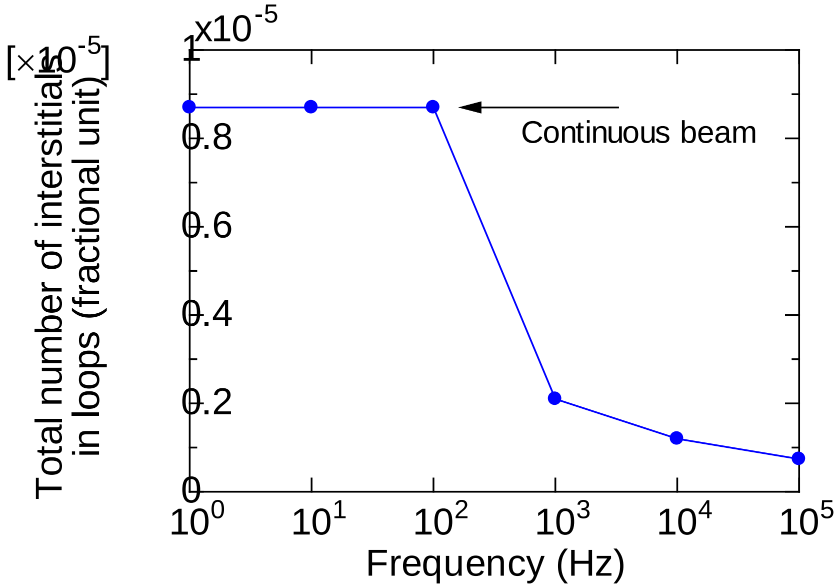

Figure 4 Total number of interstitials in interstitial type dislocation loops in Fe at the total damage of 1×10^{-2} dpa with $H = 1\times10^{-6}$ dpa/s and τ/T = 1/10 at 573 K. The arrow indicates the accumulation of interstitials under continuous beam irradiation of 1×10^{-7} dpa/s.

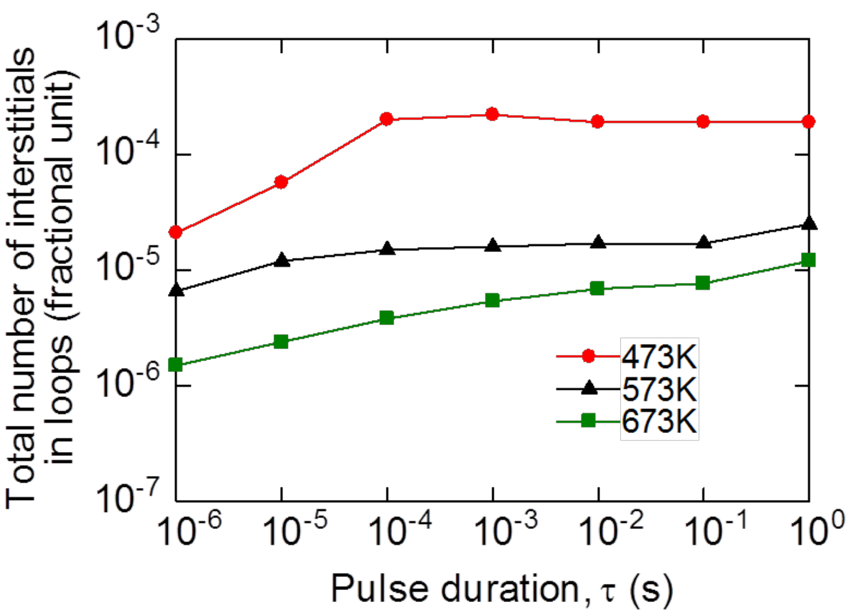

Figure 5 Total number of interstitials in interstitial type dislocation loops at the total damage of 1 dpa with $H\times\tau = 1\times10^{-6}$ dpa and T = 1sec.