# COMPUTER SIMULATION OF VOID FORMATION IN COPPER

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## ABSTRACT

In out recent experiments(1),we found that voids nucleate at vacancy clusters which trap gas atoms such as hydrogen and helium in ion- and neutron-irradiated copper. A molecular dynamics computer simulation, which implements an empirical embedded atom method to calculate forces that act on atoms in metals, suggests that a void nucleation occurs in pure copper at six and seven vacancy clusters. The structure of six and seven vacancy clusters in copper fluctuates between a stacking fault tetrahedron and a void. When a hydrogen is trapped at voids of six and seven vacancy, a void can keep their structure for appreciably long time; that is, the void do not relax to a stacking fault tetrahedron and grows to a large void. In order to explore the detailed atomics of void formation, it is emphasized that dual-beam irradiation experiments that utilize beams of gas atoms and self-ions should be carried out with residual gas atom free metal specimens.

## INTRODUCTION

When metals are irradiated with ions and neutrons at the temperature ranges of 0.4 to 0.5 $T_m$ ($T_m$ is the melting temperature), voids form which cause volume swelling and degrading of mechanical properties(2). In the materials technolo-gy of fission reactor and future fusion reactor, it is essential of develop the metals in which the formation of voids can be suppressed(3).

Growth of voids during irradiations have been reasonably understood, while the atomics of void nucleation is one of unresolved problems of the damage formation in ion- and neutron-irradiated metals. Recently we found that in neutron-irradiated pure copper the number density of voids decreases remarkably when specimens are remelted and evacuated sufficiently in ultra-high vacuum as $10^{-5}$Pa (1). Figures 1(a) and (b) shows voids that were observed by electron microscopy in (a) as-received Cu and (b) remelted Cu after neutron-irradiation at 620 K at Japan Materials Testing Reactor (JMTR). This suggests that voids are formed at vacancy clusters which trap gas atoms.

To study the detailed atomics of void formation, we carried out the molecular dynamics computer simulation which implements the embedded atom method. In the model crystal which is composed by 1372 Cu atoms, unrelaxed vacancy clusters were introduced and relaxed to the stable configuration by running a molecular dynamics at 500 K. From the results of computer simulation, we propose the nucleation model of void formation in ion- and neutron-irradiated metals.

When metals are irradiated with ions and neutrons, primary knock-on atoms are produced at the collision of projectile particle with metal constituent

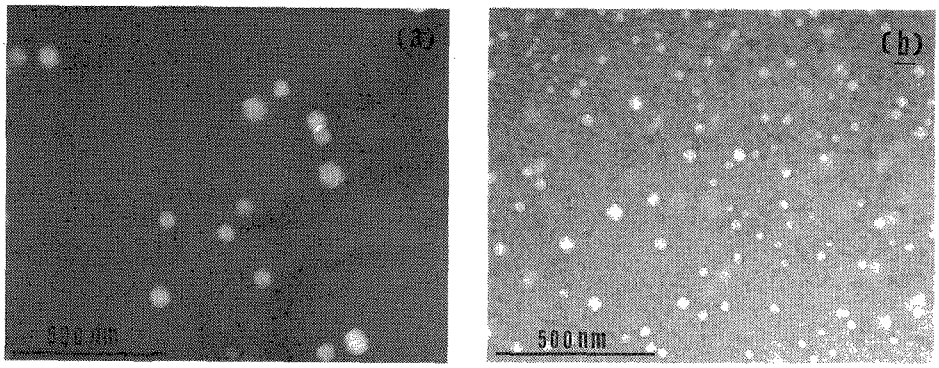


Fig. 1 Voids formed in neutron-irradiated copper at 620 K to the fluence of 5 x $10^{23}$n/m$^2$. (a) copper which is remelted and evacuated in high-vacuum before an irradiation, and (b) as-received copper.

atoms and created the displacement damage cascade. The damage cascade consists of central vacancy rich regions surrounded by interstitial clouds. At the end of cooling phase of the damage cascade formation, clusters that consist of several vacancies are formed in the core of damage cascades. This is a difference between an electron-irradiation and a particle (ion and neutron) irradiation. In an electron irradiation, vacancy clusters grow under quasi-steady state processes by absorbing a vacancy.

The objective of the present work is to propose the dual beam irradiation experiment that can examine the detailed atomics of void formation in particle-irradiated metals at high temperature.

## ATOMICS OF VOID NUCLEATION IN PARTICLE-IRRADIATED METALS DEDUCED BY MOLECULAR DYNAMICS (MD) COMPUTER SIMULATION

Results of MD computer-simulation(4,5,6) can be summarized as follows: Vacancy clusters whose sizes are below four do not relax much. The relaxation volume per vacancy is about 0.3 atomic volume for these uncollapsed vacancy clusters. An exception is a tri-vacancy. The most stable tri-vacancy is a relaxed one of Damask-Dienes type(7). The structure consists of tetra-vacancy of pyramidal shape which includes an atom in the center. This type of relaxed tri-vacancy is referred hereafter to as a 3v-sft. The formation energy of 3v-sft is 3.306 eV and almost equal to the formation energy $E_f$ of a trianglar (111) platelet. The most stable configuration of tetra-vacancy in copper is a tetrahedral void. An un-collapsed tetra-vacancy (111) platelet of diamond shape is the next stable structure.

A trapezoid penta-vacancy on (111) plane relaxes to an octahedral void which includes an atom in their center. Figure 2 shows the relaxation of penta-vacancy. The relaxed penta-vacancy is the most stable one in the size. When an additional vacancy comes to one of the nearest neighbour site of the relaxed penta-vacancy, the vacancy cluster relaxes to a stacking fault tetrahedron of hexa-vacancy (6v-sft). During a MD run, a triangular hexa-vacancy platelet on a (111) plane relaxes to a stacking fault tetrahedron. During further continuations of MD runs, a 6v-sft relaxes to an octahedral void, which is referred to as 6v-void, and relaxes back again to a 6v-sft. The structural fluctuation of hexa-vacancy between a 6v-sft and a 6v-void occurs

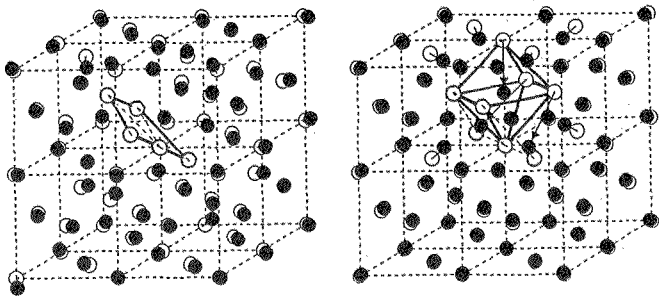

Fig. 2 The relaxation of trapezoid (111) penta-vacancy to an octahedral void that includes an atom inside a void. This relaxed structure is the most stable one of penta-vacancy.

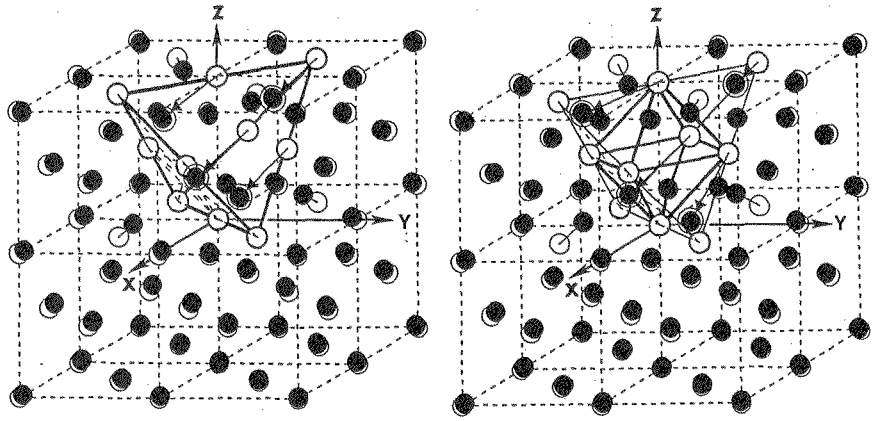


Fig. 3 Structural similarity between a stacking fault tetrahedron(6v-sft) and an octahedral void(6v-void) of hexa-vacancy. The structure consists of a tetrahedral 10 vacancy which includes four atoms inside the vacancy tetrahedron. The most stable structure of hexa-vacancy is a stacking fault tetrahedron. At high temperature, the structure of hexa-vacancy fluctuates between a 6v-sft and a 6v-void.

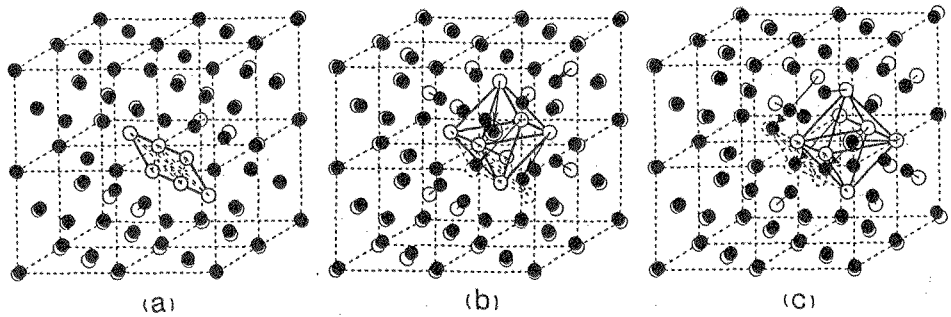


Fig. 4 A relaxation of a (111) hexa-vacancy platelet of diamond shape. During a 2 psec MD run at 500 K, it relaxed to an octahedral void which includes an atom (an encircled one) in it plus a singly vacancy. During a subsequent MD run, the whole vacancy cluster moved as shown in (b) and (c).

due to the similarity of their structures. Figure 3 shows the simlarity of structures between a 6v-void and a 6v-sft. The difference of the structure is the location of four atoms inside a tetrahedral 10 vacancy. The 6v-sft is the most stable structure in this size of clusters.

The relaxation of various configuration of hexa-vacancies was examined by MD runs. A parallelpipe hexa-vacancy cluster on a (111) plane relaxes to a relaxed penta-vacancy (Fig. 1(b)) plus a single vacancy as shown in Fig. 4.

This composite cluster of a relaxed penta-vacancy and a single vacancy moves keeping their composite structure as shown in Figs. 4(b) and (c). When these composite clusters are in the strain field due to other defects, the composite cluster can move by following the sequence as shown in Fig. 5. This kind of movement of small vacancy clusters can contribute to the coalecence of voids high fluence of ions and neutrons.

When a tetrahedral 4v-void is included in components of unrelaxed hexa-vacancy, the cluster relaxes to an octahedral 6v-void. Therefore, an octahedral 6v-void can be formed during an ion- and a neutron-irradiation at lower temperature than 0.4 $T_m$. At low temperature, an octahedral 6v-void can relax to a stable 6v-sft and does not grow to an large void. Experimentally, it was found that only stacking fault tetrahedra were observed in neutron-irradiated copper at 570 K, while only voids were observed in pure copper which was irradiated at 670 K(8). The ratio $C_{6v\text{-void}}/C_{6v\text{-sft}}$ of the concentration of 6v-void versus 6v-sft is:

$$C_{6v\text{-void}}/C_{6v\text{-sft}}$$

$$= \exp( - (E_f^{6v\text{-void}} - E_f^{6v\text{-sft}})/kT )$$

$$= \exp(-0.15\ eV/kT )$$

The calculated ratio are 0.003, 0,042 and 0.075 for 300 K, 550 K and 670 K, respectively.

When an additional vacancy comes to the nearest neighbour position of a 6v-void, the cluster relaxes to 7v-sft as shown in Fig. 6.

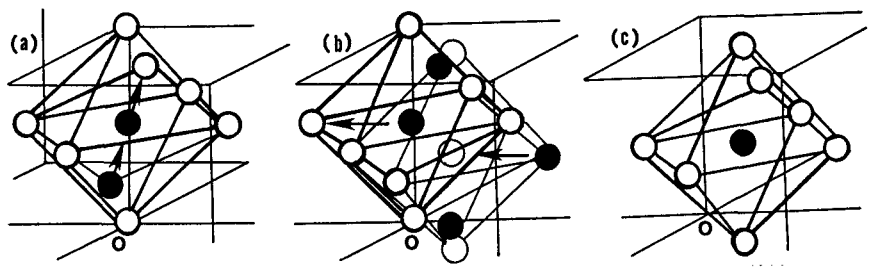


Fig. 5 Steps of movement of a composite hexa-vacancy as shown in Fig. 4(b) and (c). This kind of movement can be significant in a strain field and contribute to the growth of voids and formation of void lattice in irradiated metals.

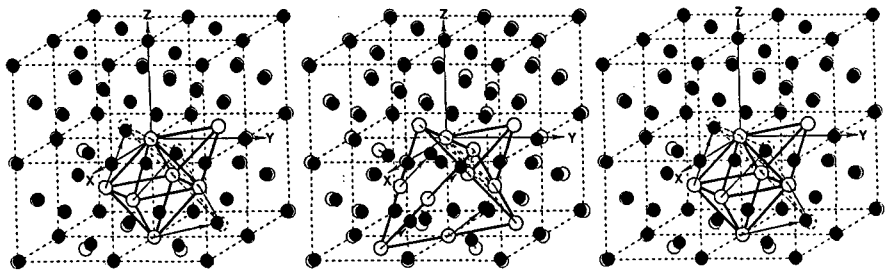


Fig. 6 Fluctuation of structure of a seven-vacancy in between 7v-sft and 7v-void.

The structure fluctuation can occur between a 7v-void and a 7v-sft.

When two additional vacancies come to the nearest neighbour position of a 6v-sft and make 8v-sft, 8v-sft did not relax to 8v-void. The cluster relaxed to a collapsed platelet as shown in fig. 7 and subsequently to a 8v-sft. When two vacancies are introduced to the nearest neighbour site of an octahedral void, a 8v-void relaxed to a 8v-sft as shown in Fig. 8 and did not relaxed to a 8v-void during subsequent a 10 psec MD run. Therefore, micro-voids can be formed by relaxing sft at the size of six and seven vacancies.

When a hydrogen comes near a 6v-void, a hydrogen atom jumps into a 6v-void. A hydrogen stays in a 6v-void for appreciably long times and jumps from vacancy site to vacancy site during a MD simulation. Figure 9 shows the caging of a single- and a di-hydrogen atom in an octahedral 6v-void during a MD run at 500 K. A 6v-void which traps a hydrogen did not relax to a 6v-sft during a 10 psec MD run at 500 K. And a 7v-void which traps a hydrogen did not relax to a 7v-sft during 10 psec MD run at 500 K. This result suggests that a gas-atom-trapped 6v-void and 7v-void can be a nucleus of voids in ion- and neutron-irradiated metals.

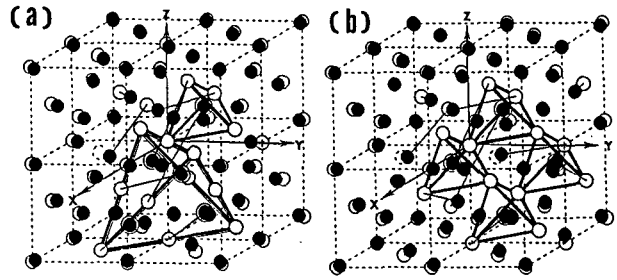


Fig. 7 When two vacancies are introduced to the nearest neighbour position of 6v-sft, this cluster relaxes into a 6v-sft plus a 3v-sft as shown in (a). It subsequently relaxed into a group of 3v-sft as shown in (b). Relaxation of a 8v-sft into a 8v-void could not be observed in the present simulation.

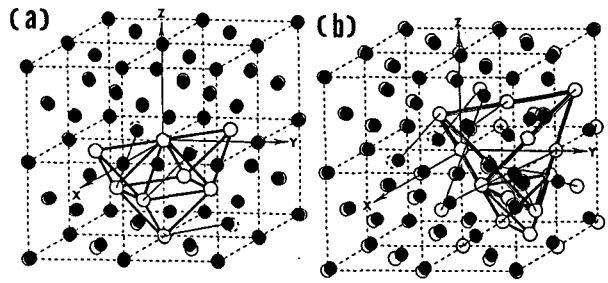


Fig. 8 When two vacancies are introduced to the nearest neighbour position of a 6v-void as shown in (a), the cluster relaxes into a 8v-sft as shown in (b): this is a 6v-sft plus a 3v-sft.

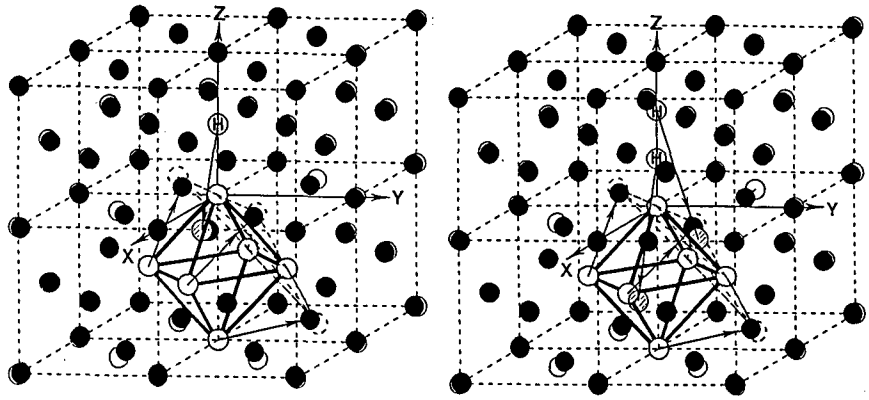


Fig. 9 When a single hydrogen and a di-dydrogen are fed into the position near a 6v-void, hydrogens immediately are trapped into a 6v-void. Hydrogen atoms move in a 6v-void and suppress the collapsing of a 6v-void into a 6v-sft. Sites where hydrogens were introduced are shown with the mark H.

## SUMMARY

The results of MD computer-simulation are summarized for the nucleation of voids in ion- and neutron-irradiated metals as follows: A single- and di-vacancy can move thermally in irradiated metals. A tri-vacancy can relax to a 3v-sft. A tetra-vacancy can relax to a tetrahedral void (4v-void). Both of a 3v-sft and a 4v-void do not move. A penta-vacancy relaxes to a configuration of penta-vacancy of an octahedral void plus an central atom can grow to a 6v-sft by absorbing a single vacancy. Both a 6v-sft and a 7v-sft can relax to a 6v-void and a 7v-void, respectively. The vacancy clusters of sft, whose size is larger than eight do not relax to void structures. These micro-voids of six and seven vacancies can be stabilized by trapping gas atoms remarkably and grow to large voids by subsequent absorption of vacancies. The present results do not deny the possibility of direct nucleation of voids at the core of damage cascades at the end of cooling phase.

Interstitial atoms which are formed at the damage cascade are ejected partially from the core by the mechanism akin to the loop punching(10). It is not definitely clear on the distribution of interstitial atoms in and near the damage cascade. Conceivable mechanism can occur to eject interstitials to outside of the core(11).

With the present nucleation model of voids in ion- and neutron-irradiated metals, the appropriate rate equations are integrated for the case of dual beam-irradiation of Cu. Results are reported elsewhere(9). Experiments of dual beam irradiation should be carried out to identify the void nucleus of six and seven vacancy clusters which trap gas atoms. It should be also examined how many gas atoms are needed to stabilize the void nucleus sufficiently.

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