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Atomistic Mechanism of Nucleation and Growth of Voids in Cu Studied by Computer Simulation

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ABSTRACT

A TEM observation of fission neutron-irradiated copper at 300°C shows that the maximum size of stacking fault tetrahedra (sft) observed is 6 nm of edge length which corresponds to a cluster of 280 vacancies and the minimum size of voids is 2.2 nm in diameter which corresponds to a cluster of 470 vacancies. The result suggests that a vacancy cluster whose size is smaller than 300 vacancies grows to sft while a cluster whose size is larger than 500 vacancies relaxes to a void in 300°C-irradiated copper. A computer simulation of molecular dynamics (MD) with an isotropic EAM potential examined this model. It is found that a vacancy cluster that is smaller than 300 vacancy segregates to a (111) platelet and relaxes to an sft. Small vacancy clusters which are generated at damage cascade cores aggregate to spherically distributed vacancies for the size of more than 500 vacancies, and relax to several (111) platelets, which finally form a vacancy (111) polyhedron. Inside a polyhedral vacancy platelet, vacancies are confined and grow to a void at high temperature.

INTRODUCTION

Shimomura and Mukouda [1] reported that a void could be formed by clustering of only vacancy. In fission-neutron-irradiated copper at 300°C to 7 x 10^{17}n/cm^2 at Kyoto University Reactor (KUR), the number density of voids exceeds the number density of hydrogen and helium atoms that are generated by the transmutation reaction [2]. A copper foil irradiated in this experiment was prepared to be a very low content of residual gas atoms by melting in highly evacuated vacuum. It is reported by the present authors that vacancy clusters move as a cluster at high temperature and coalesce to a larger cluster [3, 4]. The objective of the present work is to show how a vacancy cluster can grow to a void by clustering of only vacancies at high temperature. This means that a void can be formed without an inclusion of gas atoms in vacancy clusters in neutron-irradiated copper at high temperature. Of course if gas atoms are included in a small vacancy cluster, a void formation is promoted significantly as reported on experiments of multi-ion beam irradiated copper [5].

MODELING

Suggested Modeling on Void Formation from TEM Observations of Neutron-irradiated Copper at High Temperature

Shimomura and Mukouda [1,6] reported that both stacking fault tetrahedra (sft) and voids were observed in neutron-irradiated copper at 300°C to 7 x 10^{17}n/cm^2 in a temperature-controlled rig at KUR. The maximum edge length of observed sft was 4.5 nm (a cluster of 171 vacancies) and the minimum diameter of void observed was 2.2 nm (a cluster of 470 vacancies). We examined the maximum edge length of sft in pure copper irradiated to high fluence. In pure copper irradiated at 300°C to the fluence of 1 x 10^{20}n/cm^2 at Japan Materials Testing Reactor (JMTR), the maximum

edge length of sft was 6 nm which is equivalent to a cluster of 280 vacancies. These results suggest that a vacancy cluster whose size is smaller than about 300 vacancies relaxes to a stacking fault tetrahedron and a cluster whose size is larger than about 500 vacancies grows to a void in neutron-irradiated pure copper at high temperature.

COMPUTER SIMULATION OF MOLECULAR DYNAMICS AND MOLECULAR STATIC

A computer simulation was carried out for a copper crystal composed of 32000 atoms. The potential of the embedded atom method, EAM potential, by Daw and Baskes [7] was used to calculate the force that acts on an atom. The present EAM potential gives the stacking fault energy of 40 mJ/m^2 that is good agreement with the experimental value [8]. The periodic boundary condition was applied on crystal surface. The present author found that small strain is important on a coalescence of small vacancy clusters [3]. To include the strain effect fully, we carry out all simulation with MD. To simulate a damage evolution in neutron-irradiated copper for a long elapsed time, a MD simulation was carried out at high temperature of 1200 K. This is to accelerate a simulation with molecular dynamics (MD) technique. At first a perfect crystal was made to the thermal equilibrium at 1200K. Subsequently vacancies were introduced. In the present work, two stages of MD simulations were carried out.

In the simulation I, four voids were introduced at the positions which are apart enough with each other to prevent coalescence. The size of voids is 70 vacancies for three of them and one is a 58 vacancy. This simulation is to see that vacancy clusters whose size is smaller than 300 vacancy relaxes to sft. In the simulation II, small 31 vacancy clusters were introduced. The introduction of these small clusters is to start the simulation with vacancy clusters that were generated at displacement damage cascades. They were triangular loops and voids of which total number of vacancy is 604.

To find an intermediate defect configuration, results of MD was energy minimized by the conjugate gradient method. The vacancy was searched by counting the number of atoms in the Wigner-Seitz cell. Vacancies which locate at the nearest neighbor position were connected by a line in the figure. To identify sft, an atom alignment in crystal was observed from low index crystallographic directions as <100> and <110>.

RESULTS OF COMPUTER SIMULATIONS

Relaxation of a void of 70 vacancies to sft (Simulation I)

Fig. 1(a) shows the initial configuration of four vacancy clusters in which three consist of voids of 70 vacancies and one of 58 vacancies. After a MD run for 62 psec at 1200K, vacancy clusters of void relaxed to vacancy clusters connected as string-like by the nearest neighbor vacancy. To calculate the minimum energy configuration of vacancy clusters in this stage, a molecular static calculation of energy minimization was carried out. Fig. 1(b) shows the resultant configuration of vacancy clusters which change from voids to platelets. The structural relaxation to sft was confirmed by looking at a crystal from [110] direction as shown in Fig. 2. Sft was observed as a triangular shape. The result shows that a small cluster as 70 vacancies relaxes to sft.

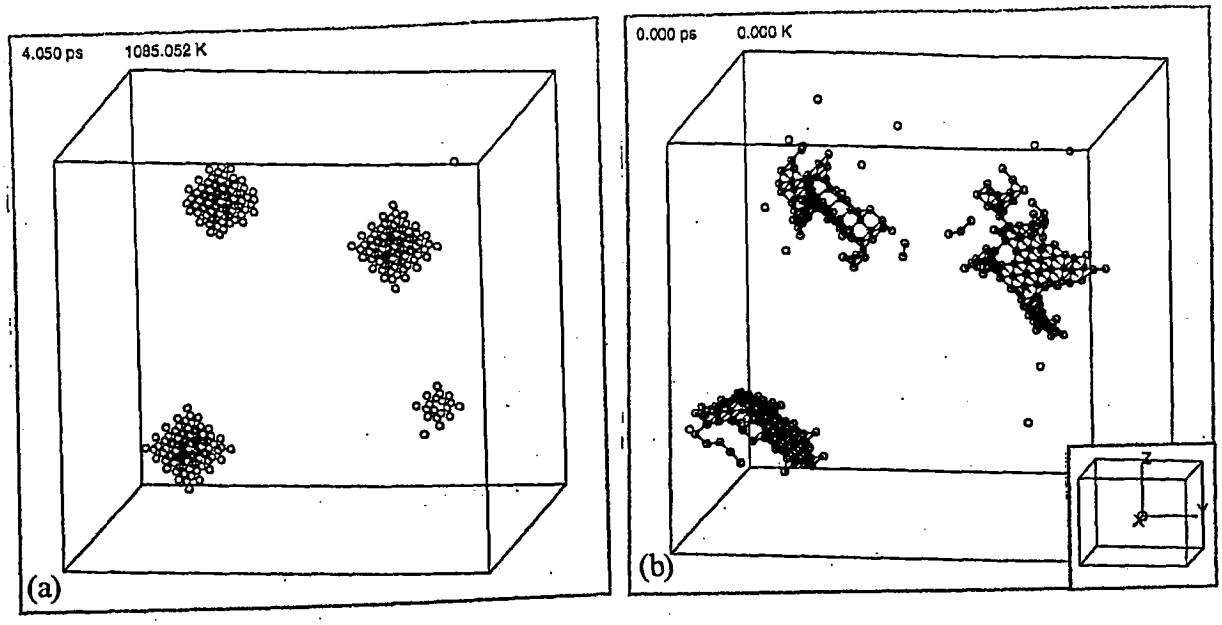

Fig. 1 Structural relaxation of four voids in copper crystal of 32000 atoms during MD at 1200 K. (a) An initial state of four voids (b) The structure of vacancy clusters after MD and the energy minimization. These vacancy clusters relaxed to {111} platelets.

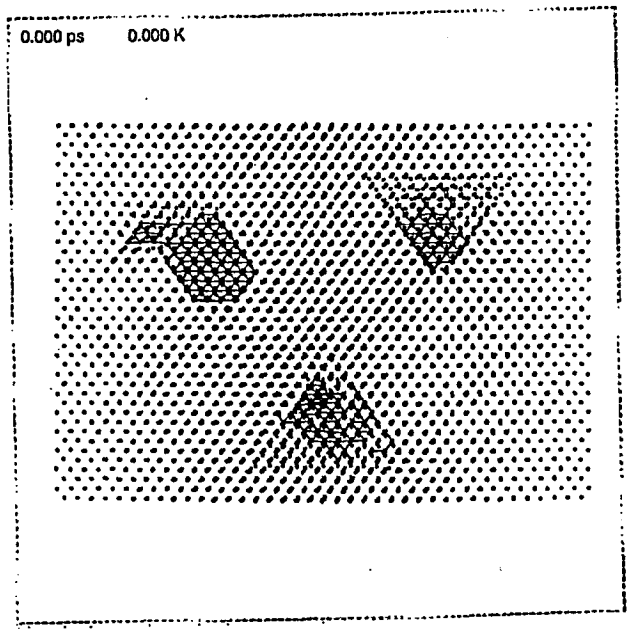

Fig. 2 A [110] projected view of crystal of Fig. 1(b). Vacancy clusters are relaxing to sft.

A vacancy cluster of 604 vacancies composed from coalescence of 31 small vacancy clusters (Simulation II)

31 small vacancy clusters were introduced as shown in Fig. 3(a). These small vacancy clusters were triangular platelets and voids. Total number of vacancies was 604. During a MD at 1200 K, these small vacancy clusters relax to string-like structures, move and unify to a large vacancy cluster with spherical distribution. After a prolong MD at 1200 K, a (111) platelet appeared clearly in a part. The vacancy distribution of this stage was energy minimized and the result was the structure not to be stacking fault tetrahedron. At edges of a large (111) platelet, generally, inclined (111)

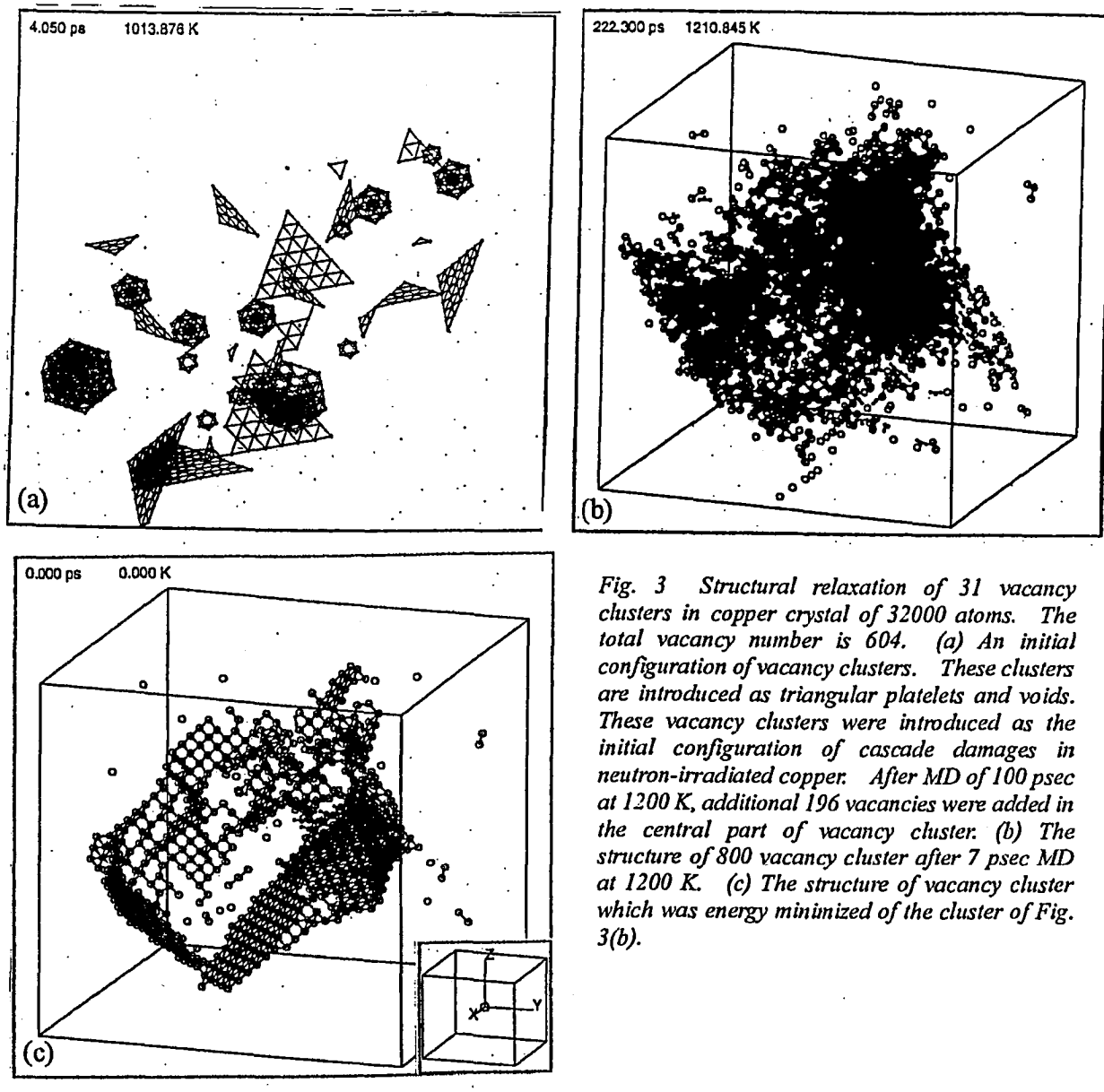

Fig. 3 Structural relaxation of 31 vacancy clusters in copper crystal of 32000 atoms. The total vacancy number is 604. (a) An initial configuration of vacancy clusters. These clusters are introduced as triangular platelets and voids. These vacancy clusters were introduced as the initial configuration of cascade damages in neutron-irradiated copper. After MD of 100 psec at 1200 K, additional 196 vacancies were added in the central part of vacancy cluster. (b) The structure of 800 vacancy cluster after 7 psec MD at 1200 K. (c) The structure of vacancy cluster which was energy minimized of the cluster of Fig. 3(b).

vacancy platelets grew. These parts prevent to relax to a whole cluster to a large sft. An evolution of a large vacancy cluster to a platelet with this inclined edge-structure limits the size of vacancy cluster to relax to sft. 196 vacancies was added to the 604 vacancy and MD was continued at 1200 K. At 220 psec MD, the distribution of vacancy was as shown in Fig. 3(b). The vacancy cluster which was obtained by the energy minimization was a vacancy polyhedron as shown in Fig. 3(c).

A Structural Relaxation of a Grown Cluster of 600 Vacancies to Vacancy Polyhedron with Confined Vacancy in It

A long term MD simulation of the 32000 atom crystal, in which 600 vacancy were introduced, were carried out at 1000 K. In this MD simulation vacancies were not introduced at an

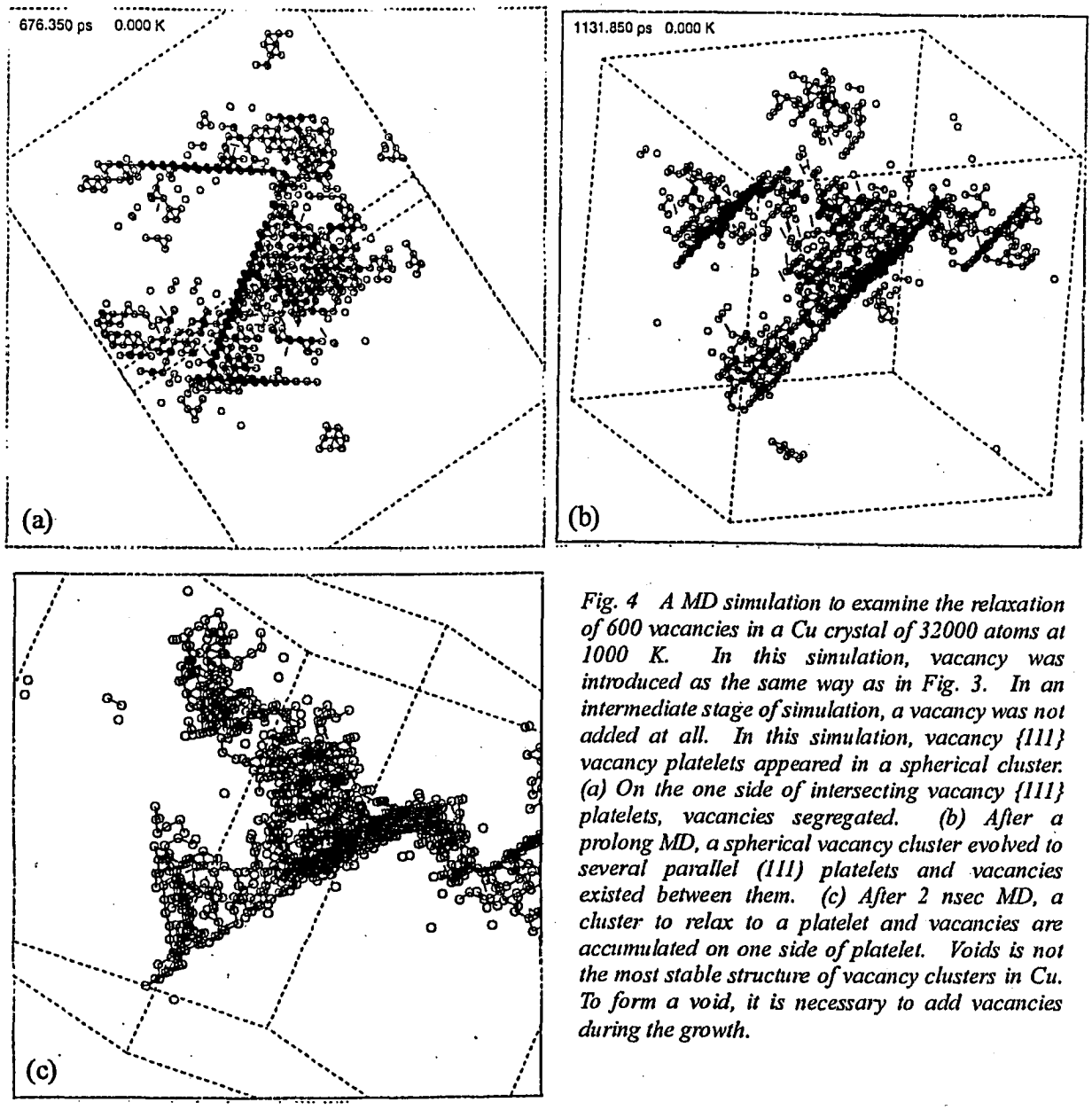

Fig. 4 A MD simulation to examine the relaxation of 600 vacancies in a Cu crystal of 32000 atoms at 1000 K. In this simulation, vacancy was introduced as the same way as in Fig. 3. In an intermediate stage of simulation, a vacancy was not added at all. In this simulation, vacancy {111} vacancy platelets appeared in a spherical cluster. (a) On the one side of intersecting vacancy {111} platelets, vacancies segregated. (b) After a prolong MD, a spherical vacancy cluster evolved to several parallel (111) platelets and vacancies existed between them. (c) After 2 nsec MD, a cluster to relax to a platelet and vacancies are accumulated on one side of platelet. Voids is not the most stable structure of vacancy clusters in Cu. To form a void, it is necessary to add vacancies during the growth.

intermediated stage. A part of vacancies were segregated to several platelets as shown in Fig. 4(a). These all platelets are not parallel, but some intersect each other. Vacancies are collected at the intersecting corner as shown in Fig. 4(b). There vacancy platelets may form a polyhedron in which vacancies are confined at the temperature of void swelling of 400°C. Voids can be nucleated at these vacancy polyhedron. A formation of such a micro-void could not be observed in this prolong simulation.

As reported by Shimomura and Nishiguchi [9], a void is not the most stable structure of vacancy cluster in copper at their size larger than 20 vacancies. The results suggest that vacancy clusters relax to the most stable configuration for a prolong MD. If vacancies are added to the cluster at an intermediate stage, they do not necessarily to relax to the structure of the energy minimum but keep a spherical distribution. A spherical vacancy cluster grows to a void during high temperature irradiation.

DISCUSSION AND SUMMARY

In neutron-irradiated copper, a vacancy cluster is produced at displacement damage cascade In a hydraulic conveyor position in KUR reactor, the main contribution of damages is due to pka whose energy lies between 10 keV and 80 keV [2]. An energy of pka which produced damages of 50 % of total damages is 40 keV. A 40 keV pka produces 100 vacancies in a cascade. In copper which is irradiated at high temperature as 300°C, vacancies move as string-like clusters and coalesce [4]. The number of cascade damage which is produced in 7×10^{17} n/cm^2 irradiation is almost the same number as ~10^{17} cm^{-3}. This indicates that vacancy clusters form at very close position for which the strain effects are important for their coalescence.

The clusters relax to sft when the size of cluster is smaller than 300 vacancies. When a cluster grows to the size of more than 500 vacancies, a cluster does not relax to sft due to the formation of inclined (111) platelet. Such a difficulty to a relaxation to sft occurs in the case if two small sft coalesce to a large cluster. These large platelets grow to a void by relaxing to a spherically distributed vacancy when additional vacancies arrive. To grow spherical vacancy clusters to voids, it is necessary to arrive additional vacancy to the cluster as in neutron irradiation at high temperature as 400°C.

In KUR fission neutron-irradiated copper at 300°C to the fluence of 7×10^{17} n/cm^2 ($E_n > 0.1$ MeV), the minimum size of observed sft was 1.3 nm which corresponds to 15 vacancy and the maximum size of sft was 4.5 nm of edge length which corresponds to a cluster of 200 vacancies. The minimum size of voids was 2.2 nm in diameter which corresponds to 400 vacancy. It was observed in *in-situ* TEM observation that a void moves as a void along <110> direction [6, 10]. A vacancy cluster whose size is larger than 400 vacancies can be nucleated by the coalescence of two vacancy clusters during neutron irradiation at 300°C. The present MD simulation was not carried out at the temperature of void swelling peak, but at high temperature. This is based on the fact that structure change with low activation energy will appear at first. Even if several different relaxation stages exist, they will appear at different MD time in the case that their activation energies are significantly different.

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REFERENCES

1. Y. Shimomura and I. Mukouda, Mat. Res. Soc. Symp. Proc., **540**, 527 (1999).
2. R. Nishiguchi, Dr. Thesis (Hiroshima University), (1991).
3. Y. Shimomura, I. Mukouda and K. Sugio, J. Nucl. Mater., **251**, 61 (1997).
4. R. Nishiguchi and Y. Shimomura, Mater. Trans. JIM., **41**, 1168 (2000).
5. I. Mukouda, Y. Shimomura, T. Iiyama, Y. Harada, Y. Katano, T. Nakazawa, D. Yamaki and K. Noda, J. Nucl. Mater., **283-287**, 302 (2000).
6. I. Mukouda and Y. Shimomura, J. Nucl. Mater., **271&272**, 230 (1999).
7. M. S. Daw and M. I. Baskes, Phys. Rev., **B29**, 6443 (1984).
8. W. M. Stobbs and C. H. Sworn, Phil. Mag., **24**, 1365 (1971).
9. Y. Shimomura and R. Nishiguchi, Radiat. Eff. Def. Solids, **141**, 311 (1997).
10. I. Mukouda and Y. Shimomura to be published in this volume.