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Computer simulation on the void formation in neutron-irradiated Cu and Ni at high temperature

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Abstract

Atomistic processes of evolution of damage structure in neutron-irradiated Cu and Ni at high temperature are investigated on the linkage of experimental results and computer-simulation. Interstitials and their clusters move to form a grouping of interstitial clusters which subsequently evolve to dislocation at high temperature. Vacancies aggregate to stacking fault tetrahedron (SFT) and void. At high temperature gas atoms do not make a significant contribution to the nucleation of voids at low fluence of neutron irradiation such as 10^{18} n/cm^2. SFT relax at a high temperature to a string of vacancy clusters, in which vacancies are connected one-dimensionally. The break-up of SFT occurs by jumping of an atom into a SFT. A loosely bound vacancy cluster of a string shape moves with low activation energy of 0.2 eV. Voids are formed by the coalescence of moving vacancy clusters of string shape at high temperature. This is due to low Helmholtz free energy of void-containing crystal than that of SFT-containing crystal. The role of gas atoms in the formation of voids is to degrade the mobility of movable vacancy clusters when captured by the clusters.

1. Introduction

Temperature controlled irradiation loop which equips a pneumatic system is developed at JMTR reactor [1]. We carried out neutron irradiations of Cu and Ni in this loop. Temperature was controlled to a constant value at 200°C and 300°C. Specimens were irradiated between 5×10^{18} and 1×10^{20} n/cm^2. Results of TEM observation can be summarized as follows [2]. (1) Interstitial clusters form their grouping [3] which develops to a dislocation. Interstitials are captured to the grouping and vacancy clusters remain uniformly distributed in specimens. Dislocations which are observed in 200°C irradiated Cu have many jogs while they are straight lines in 300°C irradiated Cu and Ni. (2) Below the temperature T_{SFT}, only stacking fault tetrahedra, which are referred hereafter to as SFT, form. The temperature T_{SFT} for Cu and Ni is 180°C [4,5] and above 250°C, respectively. The number density and the size of SFT increases with the neutron fluence. (3) Above T_{SFT}, both SFT and voids form simultaneously. The number of vacancies in a void of the average size is 350 times larger than that in the average size SFT. The number density of voids in irradiated Cu decreases with increasing neutron fluence in the range of 5×10^{18} to 1×10^{20} n/cm^2. (4) Above the temperature T_{void}, only voids form. At a low fluence such as 5×10^{18} n/cm^2, voids form uniformly in specimens while they are observed along dislocations at a high fluence. (5) The number of voids in specimens which contain residual gas atoms and in those of residual gas free is the same in specimens neutron-irradiated to the low fluence. These results suggest that the easy movement of interstitial clusters responding to strain field is responsible for the development of dislocations. The results also suggest that vacancy clusters move to aggregate a larger cluster at high temperature [6]. This is partly responsible for the decrease of void number density versus the fluence in Cu irradiated at 200°C. Computer simulation using molecular dynamics is carried out to examine the possibility of the movement of point defect clusters. Interstitial clusters move as a cluster with very low activation energy [7]. The movement is sensitive to the strain field which leads to the growth of grouping of interstitial clusters. It has been shown in the previous work that small vacancy clusters of voids and SFT relax to the structure of easy

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movement and coalesce to form a larger cluster [6]. In the present work, detailed mechanisms of void formation in neutron-irradiated Cu and Ni at high temperature are investigated.

2. Procedure of computer simulation

Computer simulation is carried out for a crystal which is composed of 4000 atoms. The size of the crystal is $10a_0 \times 10a_0 \times 10a_0$ where a_0 is the lattice constant. For the calculation of a force which acts on the atoms in the crystal, the empirical isotropic potential of embedded atom method (EAM) due to Daw and Baskes is implemented [8]. The EAM potential is parameterized by T.D. de la Rubia with the Foils' procedure [9]. The periodic boundary condition is applied on the surface of crystal. The reliability of EAM potential is checked by calculating the thermal expansion, the elastic constants, the phonon dispersion, the density of states of phonons and the energetics of point defects. The present potential reproduces these quantities reasonably. Results are reported elsewhere [10].

3. Results of computer simulation

3.1. Energetics of small vacancy clusters

The formation energy of small relaxed vacancy clusters is calculated by the energy minimization by the conjugate gradient method [11]. In this calculation, the kinetic energy of atoms is removed to be zero. Fig. 1(a) and (b) show the energy of vacancy clusters versus the number of vacancies in a cluster for Cu and Ni, respectively. In Cu, SFT is the configuration of the lowest energy. At a small size of vacancy clusters the structure is fluctuating between a SFT and a void. The structural relaxation to SFT occurs by a fundamental step of Damask–Dienes–Weizer relaxation of tri-vacancy [13]. In 3v-60 of regular triangular tri-vacancy, the change of energy during the relaxation of 3v-60 to 3v-SFT is shown in Fig. 2. In Cu, the energies of 3v-SFT and 3v-60 are almost equal and the energy barrier between these two configuration is as small as 0.02 eV. In Ni, a void is the most stable structure of vacancy clusters below the size of 30v. Nevertheless, the relaxation of 3v-SFT type occurs below the size of 30v. The energy of these relaxed structures which have 3v-SFT is higher than that of voids. The energy increment due to 3v-SFT relaxation in vacancy clusters whose size is larger than 3v is much smaller than 0.4 eV of 3v-60 relaxation. These relaxed structures appear by thermal activation. The energy difference between a void and a vacancy cluster which has 3v-SFT relaxations is not so large. The configura-

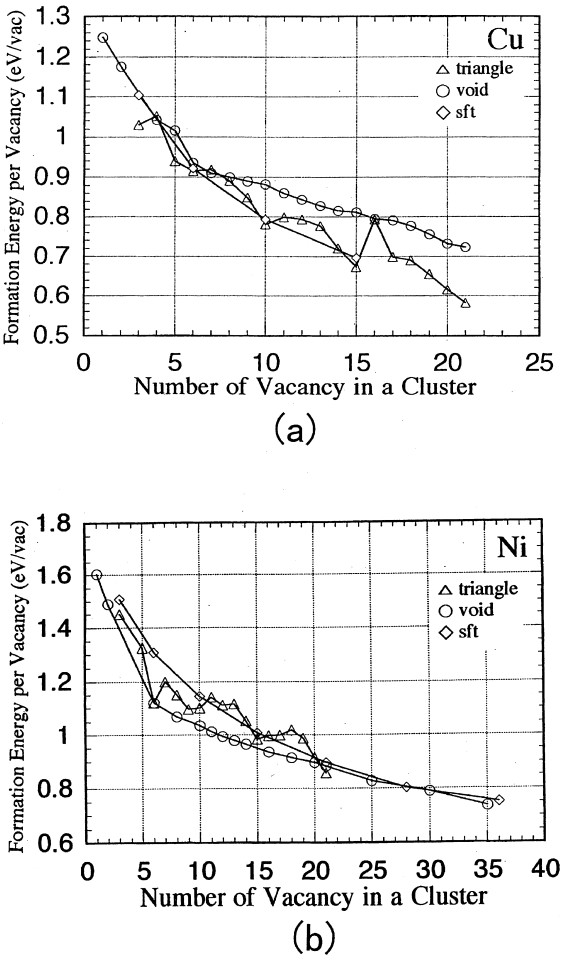

Fig. 1. The formation energy of vacancy clusters versus the number of vacancies in a cluster. The energy is calculated for a various configuration of vacancy clusters. (a) Cu and (b) Ni.

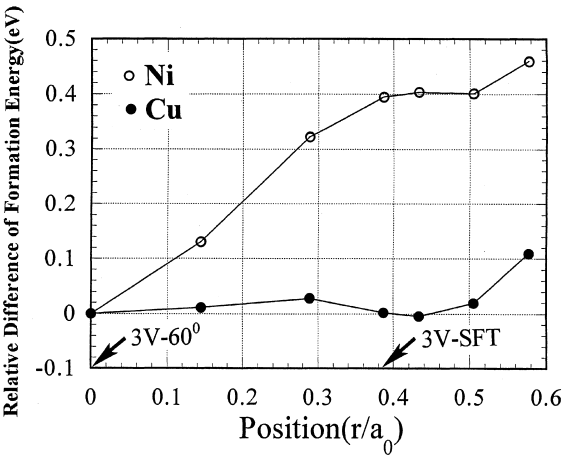

Fig. 2. The variation of energy of a 3v-60 during the relaxation to a 3v-SFT in Cu and Ni.

tional entropy is larger for a crystal which has a vacancy cluster with 3v-SFT relaxation. Due to this reason, vacancy clusters can relax at high temperature to the metastable structure of higher energy.

3.2. Dynamical behavior of vacancy cluster at high temperature

In the present work the annealing of SFT and migration of vacancy clusters at high temperature are investigated. SFT disappear at high temperature. It is found in TEM observation that SFT disappear during annealing at an elevated temperature but diminishing of the size of SFT is not observed at all. It is examined by the MD computer simulation on the atomistic process of the annealing out of SFT. Generally it is believed that a vacancy is emitted by the formation of a ledge on stacking fault plane of SFT [12]. It is found in the present computer simulation that a SFT is relaxed to a string of vacancy clusters. This occurs by jumping of one of the nearest neighbor atoms into SFT. Table 1 shows the activation energy of an atom of the nearest neighbor site to jump into a SFT. The migration energy of a vacancy cluster by keeping a cluster is also calculated as shown in Table 1. The activation energy to evaporate a single vacancy from a SFT is $E_b^{1v} + E_m^{1v}$. The value of E_b^{1v} is larger than 0.2 eV and E_m^{1v} is 0.62 eV in Cu. The activation energy of movement of a vacancy cluster is less than 0.3 eV. Therefore a vacancy cluster migrates as a whole cluster without breaking up into a single vacancy.

As mentioned previously, the vacancy cluster which is large enough to be visible by TEM relaxes to a void at high temperature. The entropy S is larger for voids due to larger number of possible configuration including relaxed structure. Free energy $H = E - TS$ of the crystal which includes a void is smaller than that of a SFT. This is the reason why we observe only voids in neutron-irradiated Ni at 300°C while we observe only a large number of SFT in Ni neutron-irradiated at 200°C.

Table 1
Activation energy of various vacancy process in Cu and Ni

	Configuration (start → end)	Activation energy (eV)	$E_f^{start} - E_f^{end}$ (eV)
Cu	1v migration	0.617	
	2v migration	0.3	
	3v-90 migration	0.227	
	relaxed 6v migration	0.106	
	3v-sft → 3v-90	0.384	0.108
	3v-90 → 3v-120	0.346	0.047
	3v-90 → 2v + 1v	0.385	0.173
	4v-twin-sft → twin-3v-90	0.208	0.045
	6v-sft → relaxed 6v	0.649	0.248
	10v-sft → relaxed 10v	0.811	0.788
	15v-sft →relaxed 15v	1.154	1.077
Ni	1v migration	1.13	
	relaxed 10v migration	0.42	
	3v → 2v + 1v	0.39	0.0

4. Discussion

4.1. The atomistic process of damage evolution in neutron-irradiated metals

When metals are irradiated with neutrons, a primary knock-on atom (PKA) is formed and displacement damage cascade is generated at the position where a PKA stops. Displacement damage cascade is generated in very small restricted volume and vacancy–vacancy interaction is very important to form vacancy clusters in the core and also interstitial-interstitial interaction is important to form an interstitial cluster in surrounding area of the core. After cooling down of the damage cascade, further clustering of point defects proceeds at high temperature. At a high temperature interstitial cluster moves by the combination of one dimensional $\langle 1\ 1\ 0\rangle$ crowdion motion and switching to another $\langle 1\ 1\ 0\rangle$ movement direction. And finally I-clusters arrive to a group of I-clusters. After removing of I-clusters, only vacancy clusters are left. I-clusters in the grouping coalesce to change to joggy dislocation loops. These dislocations evolve to complicated structure by further absorbing of interstitial clusters. At higher temperature such as 300°C, dislocations evolve to straight lines. A computer simulation has to be carried out to study the atomistic process of the evolution of dislocation structure during neutron irradiation.

Vacancy clusters relax to a collapsed structure such as SFT in Cu and Ni. They migrate as a whole cluster at a high temperature. To initiate the movement, SFT have to relax to the movable configuration of string shape. Fig. 3(a)–(d) show the relaxation of two 10v-SFT to a movable structure. The activation energy of relaxation of SFT to the movable structure is generally large and depend on the size of SFT. The activation energy of migration of string-shaped v-cluster is generally less than 0.3 eV. V-cluster movement, which has a low activation energy, sensibly reacts on the strain field. The difference of activation energy of migration to favorite direction and non-favorite direction becomes appreciable. This can be calculated from the saddle point energy during migration. V-clusters move towards the origin of strain field such as dislocations. Voids form by aggregation of such v-clusters along dislocation lines. In experiments, it was found that vacancy clusters grow to voids at the high temperature of 300°C. When two movable vacancy clusters coalesce, these grow to a void at high temperature.

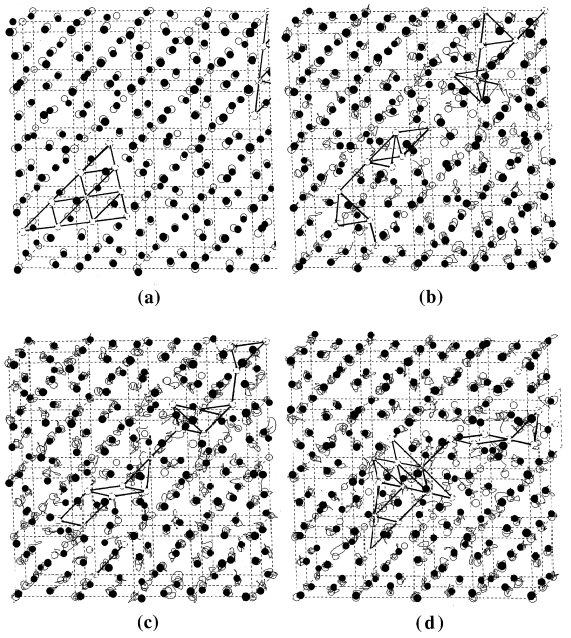

Fig. 3. The movement of vacancy clusters of string shape in Ni.

4.2. The effects of gas atoms on the formation of voids

It has been shown experimentally that gas atoms are responsible to increase the number density of voids in metals [14]. A helium atom has significant effect on the increase of the number density of voids in irradiated specimens. In our experiment [2], two different types of Cu and Ni were neutron-irradiated at 200°C and 300°C at JMTR. One type is as-received specimens which are annealed to remove dislocations at high temperature before irradiation. Another one is a residual-gas-free specimen which was melt in 10^{-8} Torr vacuum to remove dissolved residual gas. No significant difference of the number density of voids is detected in the two kinds of Cu and Ni which are irradiated to a low dose as 5×10^{18} n/cm^2. This indicates that voids nucleate at high temperature without any contribution of gas atoms. With increasing of neutron fluence, distinct difference of number density of voids is observed in between these two types of Cu samples. In neutron-irradiated Cu which is irradiated at 200°C to the dose of 10^{20} n/cm^2, we can observe remarkable difference of the number density of voids in neutron-irradiated Cu. Gas atoms can be trapped to micro-voids and degrade the mobility of voids, which leads to the void formation with high number density. Gas atoms make it difficult for small voids to move and to relax a movable string shape of vacancy cluster.

4.3. The void formation in neutron-irradiated Cu-dilute alloys

It is found experimentally that void formation can be suppressed in Cu–5%Al alloy which is irradiated to the high dose of 10^{22} n/cm^2 while difference in the number density of voids due to reducing residual gas was not observed between pure Cu and Cu–5%Ni alloy [15,16]. Generally dislocation structure does not evolve in specimens in which void formation is suppressed significantly. This means that the movement of I-clusters is suppressed remarkably by being trapped at solute atoms.

5. Summary

Atomistic processes of evolution of damage structure in neutron-irradiated Cu and Ni are investigated. The computer simulation was carried out to explore the dynamical process of evolution of damage structure and its result is discussed with the linkage of experimental results of neutron-irradiation in the temperature-controlled device at JMTR. Displacement damage cascades are formed at the positions where primary knock-on atoms stop. Interstitials and their clusters move to form groupings of clusters which develop to dislocations at a high temperature. The migration energies of interstitials and their clusters are low and the movement responds to the weak strain field. Vacancies are left after escaping of interstitial clusters to their groups. Vacancy clusters which form below the temperature T_{SFT} aggregate to stacking fault tetrahedra (SFT). At elevated temperature above T_{void}, only voids are formed. At intermediate temperatures both SFT and voids form. SFT relax to strings of vacancy clusters by jumping of an atom into SFT. The activation energy of atom jump into a SFT depends on the size of SFT. A loosely bound vacancy cluster of string shape moves with a lower activation energy of 0.2 eV. Voids are formed by the aggregation of moving vacancy clusters. At a high temperature, the Helmholtz free energy of void-containing crystal is lower than that of SFT crystal due to larger entropy term in void crystal.

References

[1] M. Narui, H. Kurishita, Y. Endoh, T. Uramoto, T. Shikama, H. Kayano, M. Kiritani, J. Nucl. Mater. 212–215 (1994) 1665.
[2] I. Mukouda, Y. Shimomura, these Proceedings.
[3] Y. Shimomura, M. Guinan, M. Kiritani, J. Nucl. Mater. 133/134 (1985) 415.

[4] S.J. Zinkle, K. Farrell, J. Nucl. Mater. 168 (1989) 262.
[5] B.N. Singh, S.J. Zincle, J. Nucl. Mater. 206 (1993) 212.
[6] Y. Shimomura, I. Mukouda, K. Sugio, J. Nucl. Mater. 251 (1997) 61.
[7] P. Zhao, Y. Shimomura, Jpn. J. Appl. Phys. 36 (1997) 7291.
[8] M.S. Daw, M.I. Baskes, Phys. Rev. B 37 (1988) 6443.
[9] S.M. Foiles, M.I. Baskes, M.S. Daw, Phys. Rev. B 37 (1988) 10387.
[10] Y. Shimomura, K. Sugio, Y. Kogure, M. Doyama, Proceeding of IUMRS, 1997.
[11] Y. Shimomura, R. Nishiguchi, Radiat. Eff. Def. Sol. 141 (1997) 311.
[12] J.P. Hirth, J. Lothe, Theory of Dislocation, McGraw-Hill, New York, 1968.
[13] A.C. Damask, G.J. Dienes, V.G. Weizer, Phys. Rev. 113 (1959) 781.
[14] K. Yamakawa, I. Mukouda, Y. Shimomura, J. Nucl. Mater. 191–194 (1992) 374.
[15] Y. Shimomura, I. Mukouda, M. Kiritani, S. Kojima, F.A. Garner, J. Nucl. Mater. 212–215 (1994) 352.
[16] I. Mukouda, Y. Shimomura, Sci. Rep. RITU A 45 (1997) 29.