An MD Simulation of the Atomistic Processes of Movement and Agglomeration of Vacancy Clusters in Nickel at 1300 K*

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Computer simulations of molecular dynamics (MD) were used to study the clustering of vacancies in nickel. This was carried out in order to investigate the atomistic processes of vacancy agglomeration in irradiated metals. Simulations were carried out using DYNAMO ver. 8.5 code, by implementing the potential of the embedded atom method (EAM). Firstly, a perfect crystal was thermalized at 1300, 1500, 1700 and 1800 K. Next, four triangular vacancy clusters whose sizes were 10 were introduced on the (111) planes at randomly selected positions. MD simulations were performed using a very small time step of outputs to observe the atomistic processes of the movement of each vacancy. It was found that the vacancy clusters moved and agglomerated whilst maintaining the individual clusters. Thus, a vacancy did not evaporate from a cluster to be absorbed into a larger cluster. There were three different processes in the agglomeration of the vacancy clusters. One was an atom caged in a tetrahedral structure of vacancies moved to another lattice site. Another was an atom caged in an octahedral structure of vacancies moved to another lattice site. The other was an atom moved along a (110) row.

(Received April 27, 2000; Accepted August 4, 2000)

Keywords: nickel, vacancy cluster, molecular dynamics simulation, embedded atom method, vacancy agglomeration

1. Introduction

When nickel is irradiated with fission neutrons at between 473 K and 500 K, vacancy clusters of stacking fault tetrahedra (sft) and voids are formed by the agglomeration of vacancies that were produced at the cores of the displacement cascade damage. The production rate of point defects is as low as 3×10^{-8} dpa/s. In the case where we investigated quenching experiments from a temperature just below the melting point, we had to cool the specimens rapidly, otherwise all of the excess vacancies were annihilated to sinks, such as the grain boundaries or dislocations. Generally it is assumed that point defects form as a small cluster after the cooling phase of the cascade damage formation. At high temperatures, the following processes are accepted as occurring in the evolution of the damage structure. Namely, a vacancy evaporates from the vacancy clusters, causing small vacancy clusters to shrink and disappear as a result, while large vacancy clusters grow to sufficient size to be observed by electron microscopy. In the present work, it is shown that another important process of movement of vacancy clusters takes a role in damage evolution.

In nickel, the formation energy was calculated using the energy minimization of the conjugate gradient method for a loop, a void and an sft.¹⁾ A void was the most stable structure for vacancy clusters smaller than 40 vacancies in size.¹⁾ The agglomeration mechanism to form these clusters was observed in a crystal introduced forty single vacancies, and we also observed small vacancy clusters agglomerating to grow into a larger cluster.²⁾ This paper describes atomistically how vacancies and clusters move in order to agglomerate.

2. Calculations

Computer simulations were carried out using the DYNAMO ver. 8.5 code, which was developed by Daw *et al.*³⁾ The embedded atom method (EAM)⁴⁾ was used, and periodic boundary conditions were applied, so there is no sink in the crystal. The size of the crystal was $10 \times 10 \times 10a_0^3$, where a_0 is the lattice constant 0.352 nm. Therefore, the perfect crystal was composed of 4000 atoms.

First, the perfect crystal was thermalized at 1300, 1500, 1700 and 1800 K by running a molecular dynamics (MD) simulation. Next, four triangular vacancy clusters whose size was 10 were introduced on (111) planes at randomly selected positions. The positions of four of the 10-vacancy clusters (clusters A, B, C and D) are shown in Fig. 1. In this figure, vacancies in the nearest-neighbor positions are connected with lines. Results were output with a time step of 0.16 ps in order to observe the atomistic processes of the movement of atoms.

Fig. 1 Four triangular 10-vacancy clusters introduced at randomly selected positions. Vacancies are connected to each other with lines. The size of the crystal is $10 \times 10 \times 10a_0^3$, where a_0 is the lattice constant 0.325 nm.

*This Paper was Presented at the Spring Meeting of the Japan Institute of Metals, held in Yokohama, on March 31, 2000.

Fig. 2 (a) The structures of three of four clusters at 1300 K after a 74.9 ps MD run. Vacancies at nearest-neighbor positions are connected with bold lines. Cluster A and cluster C agglomerated after a 101.88 ps run as shown in (b). Thin lines show every trace on which atoms moved.

Fig. 3 (a) The structures of a single cluster and a cluster agglomerated with two clusters at 1300 K after a 179.76 ps MD run. (b) Cluster A + C and cluster B agglomerated after a 194.72 ps run.

3. Results

Figure 2(a) shows the structures of three out of a total of four clusters after a 74.9 ps MD run at 1300 K. Vacancies in the nearest-neighbor positions are connected with bold lines. Cluster A and cluster C agglomerated at 1300 K after a 101.88 ps run as shown in Fig. 2(b). Thin lines show every trace on which atoms moved. Figure 3(a) shows the structures of cluster B and a cluster agglomerated from two other clusters (A + C), at 1300 K after a 179.76 ps MD run. Cluster A + C and cluster B agglomerated to form a larger cluster (A + C + B) after a 194.72 ps run as shown in Fig. 3(b). It is clear that in agglomeration, migrating atoms extended over a wide area. It was observed that as clusters moved their size was maintained, so a vacancy did not evaporate from a cluster in order to agglomerate. The agglomeration of clusters was also observed at 1500, 1700 and 1800 K.

Figure 4(a) shows the initial structure of cluster C when it was first introduced into a crystal at 1300 K. The next step is shown in Fig. 4(b) after a 2 ps MD run. During this step, an atom moved in a tetrahedral vacancy cluster and formed the structure of an sft. The larger circle shows the lattice site from which an atom moved in the tetrahedron and the line shows

Fig. 4 (a) The initial structure of cluster C at 1300 K. (b) After a 2 ps MD run, an atom moved in a tetrahedron to form a stacking fault tetrahedron. The bigger circle shows the site from which an atom moved in the tetrahedron.

the movement of the atom from the initial lattice site. Figure 5 shows the structure of cluster C after 2.98 ps MD, and also the traces of the atoms that moved during the run from the state in Fig. 4(b). It is observed that an atom caged in a tetrahedron moved to another lattice site (process-1), and the structure of the vacancy cluster is deformed. The movement of one atom was observed while another atom was moving.

Fig. 5 The movement processes of atoms during a 2.98 ps MD run.

Fig. 6 The structure of cluster C after an 8 ps MD run.

Fig. 7 The movement processes of atoms during an 8 ps MD run from their initial state. Arrows show the movement of atoms from initial to final positions.

The structure of cluster C after an 8 ps MD run is shown in Fig. 6. Figure 7 shows the processes of movement of atoms during the 8 ps MD run from the initial state of Fig. 1. Arrows on this figure show the movement of atoms from each initial position to their final locations. During this MD run, the mechanism of movement of atoms were all process-1. Thus, all the atoms were at first caged in the tetrahedral structure of a cluster and then subsequently moved to another lattice site. By this process, the cluster moved slightly.

Figure 8 shows the processes of the movement of atoms during the next MD run of 6.96 ps. The processes observed in this simulation were both process-1 and process-2 (an atom moving along a $\langle 110 \rangle$ row). It was noted that atoms moved in a sequence involving jumping onto the outer surface of a cluster, but the atoms were at first caged in a tetrahedron before jumping.

Figure 9 shows the processes of movement of atoms during an MD run timed between 46.94 and 48.94 ps. In this case, three different processes were observed, process-1, process-2 and also process-3, in which an atom was initially caged in an

Fig. 8 The movement processes of atoms during the next 6.96 ps MD run.

Fig. 9 The movement processes of atoms during an MD run from 46.94 ps to 48.94 ps. It shows the octahedral structure. Initially, the atom with a circle was caged in it.

Fig. 10 (a) The structures of clusters C and A after a 98.68 ps MD run. (b) The structure after a 99.06 ps MD run.

Fig. 11 The movement processes of atoms during an MD run from 98.68 ps to 99.06. In this figure, only a part of a cluster is shown, and process-3 movement was observed on another layer.

octahedral vacancy cluster and then moved to another lattice site. Process-2 movement was observed which extended over a longer range.

Figure 10(a) shows a part of the structure of cluster C, (upper), and cluster A, (lower), after a 98.68 ps MD run. Figure 10(b) shows a part of the same structure after a 99.06 ps MD run. The rest of the structure was on another layers.

As shown in this figure, clusters C and A had just agglomerated. The processes of movement of atoms during the MD run from 98.68ps to 99.06ps are shown in Fig. 11 and each of processes-1, -2 and -3 were observed.

The migration energy was 1.81×10^{-19} J (1.13 eV ($=E_m$)) for a single vacancy and 6.4×10^{-20} J (0.4 eV) for a small vacancy cluster (3v).¹⁾ The binding energy of the 17-vacancy cluster and a single vacancy was 1.28×10^{-19} J (0.8 eV $=E_b$). Therefore the energy for a single vacancy to evaporate from a cluster was $E_m + E_b = 3.09 \times 10^{-19}$ J (1.93 eV).

4. Summary

MD simulations were performed to study atomistic processes of four triangular vacancy clusters introduced into nickel at a high temperature. A vacancy cluster moved and agglomerated without evaporating. Three different processes were observed during the MD run. One consisted of an atom caged in a tetrahedral structure of vacancies moving to another lattice site. Another was an atom caged in an octahedral structure of vacancies that moved to another lattice site, and the final process involved an atom moving along a $\langle 110 \rangle$ row. The clusters moved to agglomerate during these three processes.

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