TEMPERATURE EFFECTS ON THE PRIMARY STATE OF DISPLACEMENT CASCADE

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ABSTRACT

The effect of lattice temperature on the primary state of displacement cascade damage is investigated by using molecular dynamics technique. Number of atoms are large enough to prevent all the displaced atoms from escaping from the calculated region, and the calculated region is surrounded by periodic boundary condition for making thermally insulated system. Cascade damage equilibrating processes are discussed. It is shown that temperature of the target material affects both the cascade cooling processes and the initial defect configuration. It is suggested that damage parameter function must include irradiation temperature of the target materials. Influences of the target temperature on the cascade collapse are also discussed.

Introduction

In the materials used in an irradiation environment as nuclear fission and nuclear fusion reactors, irradiation-induced displacement cascade generates high density of lattice defects. The behavior of these defects is considered to be one of the most important factors for property degradation of such materials. In order to evaluate the lifetime of materials in a nuclear fission reactor or to design materials for a nuclear fusion reactor, establishing correlation between the irradiation dose and the degradation of materials is required. However, such correlations have not been clarified well so far. There are two steps to establish such correlations; one for the correlation between irradiation dose and lattice defects produced by the cascade damage, the other for the correlation between irradiation induced lattice defects and the property change of the materials. In the latter case, not only the number of defects, but defect structures are required for better description of the correlation. The number of displaced atoms (displacements per atom; dpa) has been often used as a unit of dose. Nevertheless, it is not a best parameter for

describing the correlation. In other words, such information should be obtained for establishing dose-property correlation as the number of defects, clustering probability of defects of each type, the fraction of mobile or immobile defects and configuration of the defect clusters.

Cascade damage evolution processes are classified into the following five stages; collisional phase, relaxation phase, cooling phase, thermal phase and diffusional phase. Incident particles displace some atoms, which are called 'primary knock-on atoms; PKAs'. The PKAs also produce secondary knock-on atoms. Such sequential collisions occur during the collisional phase. During the following relaxation and cooling phases, displaced atoms are relaxed and deposited energy is dispersed, respectively. After the system becomes thermally equilibrated, defect interactions occur inside and outside of the cascade region during the thermal and diffusional phases, respectively. While thermal and diffusional phases are both equilibrium processes, collisional, relaxation and cooling phases are non-equilibrium processes so that the physical treatments are not so easy. Let us take the defect clustering for example. The cluster formation during the thermal and diffusional phases are regarded as the problem of the defect jump frequency and thermodynamic stability. On the other hand, the vacancy cluster formation during the cooling phase, which is called 'cascade collapse', was considered to be the result of thermal gradients within the cascade or due to drift under the influence of the strain field of the growing cluster [1][2]. More recently, Averback et al. have shown that the vacancy cluster can be formed by resolidification of molten cascade core in an epitaxial fashion as described below[3]. Molecular dynamics method is considered to be one of the most useful techniques to investigate such non-equilibrium phenomena.

Molecular dynamics technique has provided us some important information about cascade damage processes; cascade processes are controled by two phenomena [4] [5], i.e., replacement collision sequences during the collisional phase and liquid-like structure at the cascade core during the cooling phase [6]. Interstitials, which are away from the molten core of the cascade via replacement collision sequences, are able to escape from spontaneous recombination with vacancies and survive. In addition, during the resolidification of the molten cascade core, an interstitial prismatic dislocation loop has been observed to be generated at the solid-liquid interface [7]. Moreover, when the cascade occurs near the pre-existing defect clusters, defect survival ratio is decreased by the cascade interaction [8]. These phenomena can not be seen by experiment, and it is an advantage of molecular dynamics technique. However, because of the computer simulation, there are limitaions as to the number of atoms to be incorporated in the calculation and to the time scale to be handled by this method. Therefore, alternative method [9] has been tried to study the cascade from the macroscopic point of view [10]

[11]. However, quantitative data have not been obtained yet.

In this paper we show the results of molecular dynamics simulation of cascade damage processes, and discuss the temperature effects on the initial defect configuration and on the cascade cooling processes, considering the cascade collapse and the better description of damage parameter function.

Computational methods

Cascade damage evolution is so short (~10 psec) and takes place with in such a small volume (~10^{-26} m^3) that it is very difficult to investigate the processes by experiments. Therefore, to study the cascade processes, we use molecular dynamics computational technique in which the equations of motion for all the atoms in the calculated region are solved.

Cascade damages are calculated by molecular dynamics technique. They are initiated from a 250 eV PKA in fcc gold at such initial temperatures as 0, 100, 300 and 1000 K. The total number of atoms used in these calculations is 10800, and the periodic boundary condition is adopted to make thermally insulated systems. Interatomic potential used here is a simple embedded atom method (EAM) potential of Oh and Johnson [12]. $3x10^{-15}$ sec is taken as a time step for the calculation.

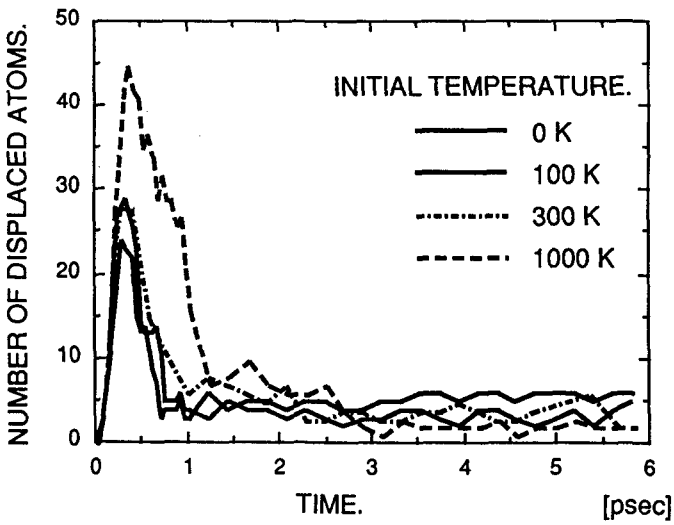

Figure 1. Time dependence of the number of displaced atoms at various initial temperatures.

Results and discussion

Figure 1 shows time dependence of the number of displaced atoms for four initial temperatures. A displaced atom is defined to be displaced by over one-quarter of a lattice constant from nearest lattice site. During the collisional and relaxation phases, the number of displaced atoms are rapidly increasing and decreasing, respectively. The number of displaced atoms at the end of the collisional phase is larger for higher initial temperature, while during the cooling phase more displaced atoms are recombined spontaneously with the opposite type of defect at the higher initial temperature.

The number of displaced atoms at the end of the collisional phase (displacement per atom: dpa) is currently used as 'damage parameter function', which has been used as an exposure parameter and sometimes as a correlation parameter between the irradiation-induced lattice defects and the material property changes. Dpa is calculated from irradiation fluence, cross section of collisions and damage energy, but it does not incorporate the effects of temperature of the target materials. Figure 1 indicates that the temperature effects should be included into

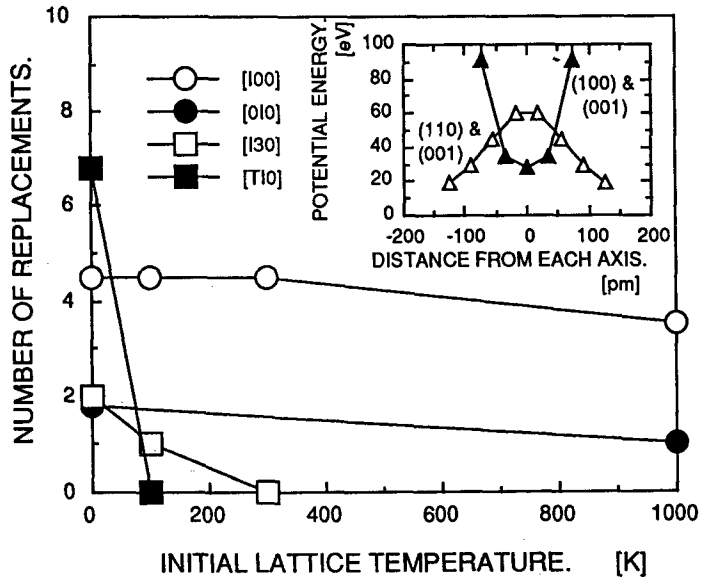

Figure 2. Temperature dependence of length of replacement collision sequences along various crystallographic direction. An inset in this figure shows potential energies on the intersection of two planes; one is (001) plane and another is the plane perpendicular to the [110] or [100] axis at the center of two atoms along each axis.

the damage parameter function. Temperature effects may be incorporated in the form of the temperature dependence of 'damage parameter efficiency', which is defined as the ratio of the final defects to the damage parameter function. However, the damage parameter function as given by the NRT model , for example, does not include the temperature effects during the collisional phase. Temperature effects before the end of the collisional phase and after the end of the collisional phase should be separated with each other. The latter effect is mainly related with the interactions of defects, while the former effect is related with the displacement of atoms by the thermal vibration as mentioned later.

At this energy range (250 eV), the collision sequences along close-packed directions are dominant to dictate the damage structures. Figure 2 shows temperature dependence of the length of replacement collision sequences (RCSs) at the end of the collisional phase. The length becomes shorter at higher temperature, and moreover, there is a directional dependence of the temperature dependence. <110> direction is strongly affected by the lattice temperature, while the length of RCSs along <100> direction is almost unchanged. Total energy changes are calculated by the density functional method [13] when one atom is moving along the <110> and <100> axes. An inset in figure 2 shows the total energy distributions when one atom is moved on the two intersections; one is that of (110) and (001) planes, and the other is that of (100) and (001) planes. These curves are considered to show the potential energies of the atom moving on <110> and <100> axes. Because of computational limitations, copper atoms are used in this calculation instead of gold, but it may not be unreasonable to think that the major conclusion may still hold for gold since the nature of RCSs is considered to be

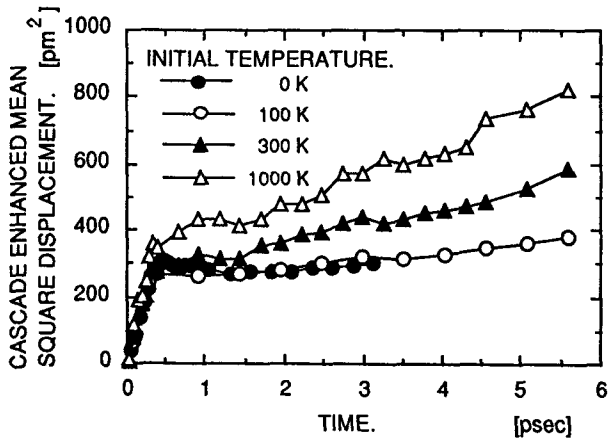

Figure 3. Time dependence of cascade enhanced mean square displacement at various initial temperatures.

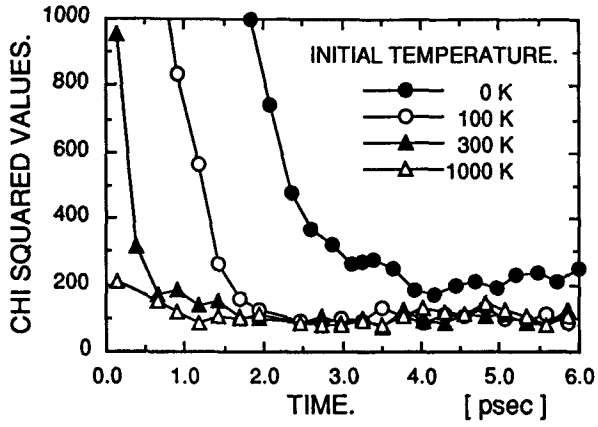

Figure 4. Time dependence of chi squared values representing the difference of velocity distribution and Maxwellian distribution at various initial temperatures.

strongly affected by the crystallographic structure. Moreover, relaxation of ambient atoms is neglected, because the velocity of atoms in RCSs during the collisional phase is so large that the relaxation is not so prominent. Along the <100> axis the potential curve has a local minimum, while along the <110> axis it does not show a local minimum. Namely, the atoms moving along the <110> axis easily slide down the potential curve because of the fluctuation of the potential curves by the thermal vibration. On the other hand, the atoms along the <100> axis are moving within the trough of the potential surface so that the RCSs along <100> are considered to be stable for thermal vibration.

At higher temperature the length of RCSs becomes shorter and the relative distances between vacancies and interstitials become shorter. This may affect the interaction between vacancies and interstitials after the relaxation phase.

Although the length of the RCSs becomes shorter at higher temperature as shown in figure 2, the number of displaced atoms is increasing with increasing temperature as shown in figure 1. This comes from the difference of energy branching in the cascades at different temperatures. Figure 3 shows the time dependence of the net mean square displacements (MSDs) enhanced by the cascade damage processes, which do not include the MSDs induced by the thermal vibration. MSDs are larger for higher temperature. Therefore, the number of excited atoms than normal thermal vibration is increasing with increasing temperature, namely, at higher temperature energy branching becomes more active so that the number of collisions between atoms is expected to increase with temperature. This implies that cascade processes are equilibrated quicker at higher temperature. This expectation is supported by figure 4, which shows the time de-

pendence of chi squared values representing the degree of differences between the velocity distribution for all atoms in the calculated region and the Maxwellian distribution. Maxwellian distribution is not the strict solution of equilibrium state of condensed matter, but it may be a good approximate solution.

C. English and M. Jenkins have described in their paper [2] that cooling rate at the cascade core is decreasing with higher temperature by the classical thermal model as results of temperature gradient and of the change in the lattice thermal conductivity. This assumption does not agree with our results. Our calculational model is for thermally insulated solid and is of quite low energy cascade so that we cannot make a simple comparison. However, our results suggest that cooling rate of the cascade core is strongly affected by the collisional processes during the collisional phase (energy branching, defocusing of collision sequences). Moreover, temperature effects on the collapse probability is related, not only to the cooling rate of the cascade core, but also to the initial defect configuration. At higher temperature initial vacancy configuration becomes more compact and distances between vacancies and interstitials are closer. These factors are considered to play an important role for the effect of temperature on the cascade collapse probability.

Conclusion

We have discussed the temperature effects on the cascade damage evolution processes, in particular on the cooling processes of the cascade. The effects indicate the defocusing by the fluctuation of the potential surface by the thermal vibration and the different equilibrating processes with different energy branching. These phenomena during the collisional and cooling phases are found to be strongly dependent even on the small fluctuation of lattice atoms by the thermal vibration.Therefor it is quite natural to think that they are dependent on the crystallographic structure of metals. In fact, cascade collapse probabilities strongly depends on the crystallographic structure [2]. In other words, the efficiency of thermal dispersion via replacement collision sequences and their defocusing may play a key role for the defect interaction during and after the cooling phase including cascade collapse.

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References

[1] V.I.Protosov and V.G.Chudinov: Rad.Eff. 66, 1(1982).

[2] C.A.English and M.L.Jenkins: Mater.Sci.Forum 15-18 (1987) pp.1003-1022.

[3] R.S.Averback, T.Diaz de la Rubia and R.Benedek, Nucl.Inst. and Methods in Phys.Res. B33, 693(1988).

[4] T.Diaz.de la Rubia, R.S.Averback, R.Benedek and W.E.King: Phys.Rev.Lett. 59,17, 1930 (1987).

[5] R.S.Averback, H.Hsieh, T.Diaz de la Rubia and R.Benedek: J.Nucl.Mater. 179-181, 87 (1991).

[6] C.A.English and W.J.Phythian and A.J.E.Foreman: J.Nucl.Mater. 174,135 (1990).

[7] T.Diaz de la Rubia and M.W.Guinan: Phys.Rev.Lett. 66,21, 2766 (1991).

[8] W.J.Phythian: to be published in Rad.Eff.

[9] M.Caro, A.Ardelea and A.Caro: J.Nucl.Mater. 183, 33 (1991).

[10] K. Morishita, N.Sekimura and S.Ishino: to be published in Rad.Effects.

[11] K.Morishita, N.Sekimura, T.Toyonaga and S.Ishino: Proc. of ICFRM-5, to be published in J.Nucl.Mater.

[12] D.J.Oh and R.A.Johnson: J.Mater.Res.3,3, 471 (1988).

[13] P.Hohenberg and W.Kohn: Phys.Rev. 136,3B, B864 (1964).