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Evaluation of Threshold Displacement Energy in Tungsten using Molecular Dynamics Simulation

Mosab Jaser Banisalman, Takuji Oda*
Department of Nuclear Engineering, Seoul National Univ., 1 Gwanak-ro, Gwanak-gu, Seoul 151-742
**Corresponding author: oda@snu.ac.kr*

1. Introduction

In nuclear materials, collision of constituent atoms with energetic particles such as high-energy neutron and atoms cause defect formation due to atomic displacement. Formed defects often cause adverse effects on material properties, such as irradiation induced hardening and a shift of the ductile to brittle transition temperature to higher values [1]. In order to control and mitigate adverse radiation effects, it is important to understand how radiation defects are formed and evolved in materials during reactor operation.

The threshold displacement energy (E_d) is the minimum recoil energy that a lattice atom needs to be displaced to an interstitial position in a material, thus forming a stable Frenkel pair. It can be used to determine the number of defects created by energetic particles [2]. Therefore, E_d is regarded as one of the most fundamental quantities in determining the primary state of radiation damage in materials [3].

Tungsten (W) is a promising candidate material for plasma-facing components in fusion reactors. Since plasma-facing components are used under irradiation of 14 MeV neutron and plasma particles, radiation damage processes and its effects on material properties need to be understood. However, there is some inconsistency in E_d used in previous studies: some studies use a value around 45 eV, and others 90 eV. Since this large difference in E_d cause a large difference in the expected number of defects formed in W, it is needed to find an appropriate value for E_d.

Hence, in the present study we utilize molecular dynamics (MD) method to determine E_d of W. There are several calculation settings that affect the accuracy of the E_d determination. In the present work, we especially focus on the effect of system size. Specifically, we analyze how the system size changes the average E_d ($E_{d.avg}$), which is the effective threshold displacement energy obtained by taking average of E_d of various recoil directions. We also analyze how recoil direction affects E_d value in W.

2. Methods

All MD simulations are performed using the LAMMPS code [4]. The interatomic interactions are described by an embedded-atom method (EAM) potential, which was originally developed by Murray Daw and Mike Baskes and further improved by C. Björkas and K. Nordlund [5] on the repulsive and the electron density function. [6].

In order to avoid overlapping the damage region and reentered recoil atoms along <111> collision sequence, we avoid using a cubic system. Instead, orthorhombic supercells with periodic boundary conditions are used: 2×2×4, 4×4×6, 6×6×9, 8×8×12, 10×10×15 and 18×16×14. Before initiating a recoil event, the simulation cell is equilibrated with 30 K and 0 Pa, which is used as the initial conditions of all simulations. In each recoil event, simulation of around 5 ps is conducted. An adaptive time step is used with a maximum displacement (x_{max}) of 0.01 Å per step and the maximum time step (t_{max}) of 0.002 ps. We confirmed that this setting is accurate enough for E_d evaluation in comparison with several other x_{mas} and t_{max} settings.

A recoil MD simulation is performed by giving a recoil energy to an atom located at around the lattice center, which is regarded as primary knock-on atom (PKA). The recoil energy is converted to velocity components of PKA when it is given to PKA using the following equation.

$$[\text{recoil energy}] = \frac{1}{2}Mv^2 , \qquad (1)$$

where M is mass and v is velocity of PKA.

Starting with 10 eV, we increase the recoil energy by 6 eV until defects are firstly detected. Then we decrease the recoil energy by 1 eV to reach the minimum energy for defect formation, which is defined as E_d in the present study. A reliable value for the average threshold displacement energy ($E_{d,ave}$) is calculated for selected 100 isotropic displacement directions and for 100 different recoil-event timings.

3. Results

3.1 Direction effect for E_d evaluation.

Defects formation mechanisms largely depend on projection direction. As examples, we pick up specific <100> and <321> projections to show typical recoil events in this section.

The recoil energies 10 eV, 100 eV, 175 eV and 200 eV were chosen to study defects formation probability for <321> projection. System structures obtained after the recoil event (5 pa after giving a recoil energy to PKA) are compared in Figure. 1. Defects were formed with 175 eV and 200 eV, while not with 100 eV and 10 eV.

The potential fields of the atoms in the lattice form a barrier over which PKA must pass in order to be displaced. This is the source of the displacement threshold energy. If the recoil energy is less than E_d, PKA only vibrates around its equilibrium position and does not be displaced. This vibration of PKA is transmitted to neighboring atoms through the interaction of their

potential fields and the initial kinetic energy of PKA are finally changed to heat.

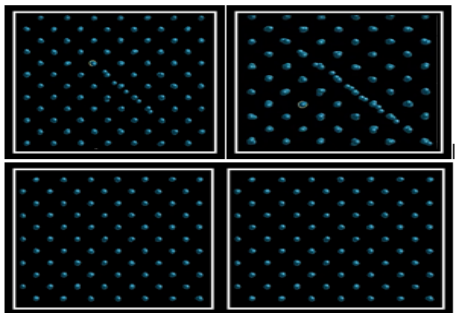

Fig. 1. System structures at 5 ps after collision event for simulations of four recoil energies for <321> projection. The recoil energies are 200 eV, 175 eV, 100 eV and 10 eV from upper left to lower right.

If the recoil energy is greater than E_d, it may be reasonable to assume that the defect number increases with the recoil energy; however, this assumption is not necessarily correct. Figure 2 shows the probability for defect formation as a function of recoil energy for <100> projection. We see that contrary to the assumption, there are no defect production at some recoil energies for <100> projection even it is larger than E_d. For example, in Figure 2, although defects formed at 43 eV, which is E_d, there are no defects formed at 100 eV.

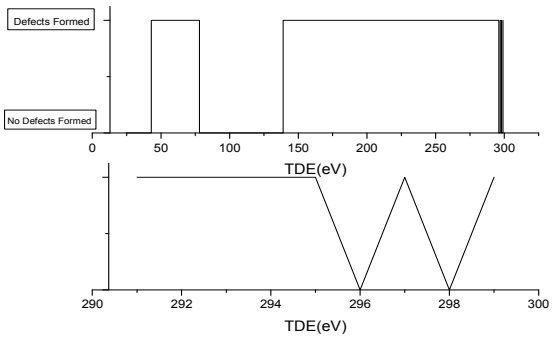

Fig. 2. Probability to form defects for <100> projection as a function of recoil energy. E_d is determined to be 43 eV in the upper panel.

In fact, there are directions in which the surrounding atoms will remove large amounts of energy from PKA. In this case, as a large fraction of the recoil energy is immediately converted to heat, a larger recoil energy is required to displacing PKA. For example, a large E_d value, 173 eV, is found for <321> projection. On the other hand, along high symmetry directions, there exist open paths along which the number of atoms colliding with PKA is small. In this case, since the recoil energy is effectively used for defect formation, E_d is lowered. For example, E_d for <100> projection is just 43 eV.

We obtained 43 eV, 173 eV, 160 eV, 145 eV, 90 eV, and 55 eV as E_d for <100>, <321>, <3.1 7.4 11.3>, <563>, and <1.1 0.2 0.3> projections, respectively.

The more indices the displacement direction has, the less symmetry the collision pattern in lattice system shows. With less symmetry the recoil particle has higher probability to collide with neighboring atoms. This causes loss of its kinetic energy, and then a larger recoil energy is needed to overcome the potential barrier for displacement.

Another possible reason for the strong direction dependence of E_d is that the potential barrier surrounding an equilibrium lattice site is not uniform in all directions in crystallographic metals [2]. For some directions, a larger energy is needed to displace PKA than others.

3.2 Size effect for $E_{d,\,avg}$ evaluation.

Size effect has direct influence for the E_d evaluation. With a larger system, a more accuracy in calculation is expected. If the system size is too small, the system temperature after the recoil event becomes high as the recoil energy is converted to thermal energy, which increase the thermal recombination effect on the formed defects. Defects-defects interactions in neighboring MD images can be a source of another system size effect on the E_d evaluation. However, with the acceptable value of error in E_d, a proper system with least number of atoms can be taken. We can be away from the thermal recombination effects choosing proper selection criteria.

In Figure 3, $E_{d,\,avg}$ determined in systems of different sizes are compared. We evaluated $E_{d.avg}$ with increasing the system size from smaller to larger number of atoms. 18×16×14 is the maximum tested size and has 94 eV of $E_{d.avg}$. $E_{d.avg}$ converges to a constant value around 95 eV as the system size increases. This result suggests that 90 eV is a more appropriate value for E_d of W, rather than 45 eV used in some previous studies. 95 eV is the recommended value for $E_{d,ave}$ of W by the present study.

In Figure 3, when the system size is smaller than 4×4×6, calculation results show a divergence. This is because the system temperature becomes larger than 3695 K, which is the tungsten melting point temperature, after the recoil event in 4×4×6 system. When the system is melt, atoms are not necessarily located around lattice sites. Then, we cannot appropriately detect lattice defect and cannot determine E_d.

The system temperature after the recoil event given in Table 1 can be used as a criterion to determine a proper system size with respect to the thermal recombination effect of formed defects. The final temperature of the MD simulation at 5 ps is approximately expressed as following equation.

$$\frac{1}{2}E = \frac{3}{2}Nk\Delta T \quad , \quad (2)$$

where E is the recoil energy, N is the number of atom, k is Boltzmann constant, ΔT is difference between initial and final system temperature. For example if $E_{d.avg}$ = 100 eV, the final temperature in 4×4×6 supercell is approximately 2300 K.

Table 1: Average E_d for different system sizes averaged over 100 different recoil-event timings and 100 isotropic displacement directions. Temperatures given in the table are

obtained by averaging the temperatures at 5 ps in simulations where the recoil energy was equal to E_d.

Dimension	Atoms	Ed (eV)	Temp (K)
2×2×3	24	62.5075	8500
4×4×6	192	147.7325	2830
6×6×9	648	105.7375	590
8×8×12	1536	102.005	270
10×10×15	3000	96.89	150
18×16×14	8064	94.7375	71

The temperature criteria should be set according to the probability of defect recovery within the recoil simulation period (5 ps). A previous study of MD simulation on collision cascades in W shows that the number of defects decreases by 20% from 300 K to 1025 K in W [7]. This result means that defect recovery largely occurs between 300 K and 1025 K. In this temperature range, although the vacancy migration rate is still low, self-interstitial atoms start migrating very fast so that primary formed self-interstitial atoms has higher chance of recombination with vacancies. To avoid thermal defect recovery after the recoil event, we conservatively set a temperature criterion that the temperature at the end of the recoil event should not go beyond 300 K to avoid the thermal recombination of defects after the event. Among the results in Table 1, 270 K is the first temperature lower than our criterion temperature. This system gives 102 eV as $E_{d,avg}$, which differs from the converged value (~95 eV) by around 5%. If the required accuracy in E_d is 5%, 8×8×12 is an appropriate system size to minimize the calculation cost.

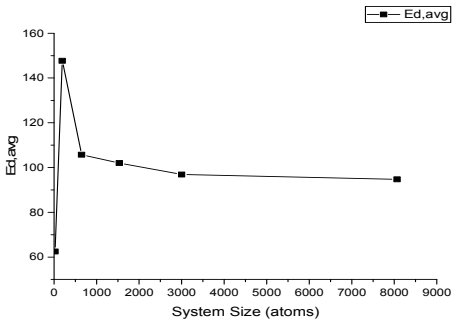

Fig.3. $E_{d,ave}$ as a function of system sizes.

4. Conclusions

We determined the threshold displacement energies of W using MD calculation. We first evaluated E_d for specific projections. The largest E_d value (173 eV) is obtained for around <321> projections, and the smallest E_d value (43 eV) for around <100> projections. We assigned the reason of the large direction dependence of E_d to direction dependences of (1) the potential energy barrier for displacement and (2) the number of collisions between PKA and other atoms, which determine how effectively the displacement energy is used for defect formation.

For the determination of $E_{d,ave}$, E_d over 100 directions with 100 different recoil-event timings were calculated, and then averaged. In a comparison of $E_{d,ave}$ values calculated with systems of different sizes, we found that $E_{d,avg}$ converges to a constant value around 95 eV as the system size increases. This converged value is the recommended value for $E_{d,ave}$ of W.

A smaller system size is preferable to reduce the calculation cost. However, for achieving a required statistical accuracy, some criteria should be satisfied when a small system is employed. In the present study, we focused on thermal defect recovery effect after the recoil event as a possible source of system size effects. We set a selection criteria that the system temperature after the recoil event should not go beyond 300 K. 8×8×12 system agrees with this selection criteria, which gives $E_{d,\mathrm{avg}}$ within 5% error from the converged value.

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