M&C 2017 - International Conference on Mathematics & Computational Methods Applied to Nuclear Science & Engineering, Jeju, Korea, April 16-20, 2017, on USB (2017)

Evaluation of Threshold Displacement Energy of Tungsten by Molecular Dynamics Simulation

Mosab Jaser Banisalman[*1], Sehyeok Park, Takuji Oda[*2]

Department of Nuclear Engineering, Seoul National University:
1 Gwanak-ro, Gwanak-gu, Seoul 151-742, South Korea
*[*1] mosab123@snu.ac.kr, [*2] oda@snu.ac.kr*

Abstract - *Threshold displacement energy (TDE) is an important quantity to determine the number of defects formed by irradiation of high-energy particles. For tungsten, different values of TDE have been reported in several studies, which caused discrepancy in calculated damage amounts. In the present study, we evaluated TDE using molecular dynamics simulation, where TDE is defined as the average value of minimum displacement energies to create a stable defect over all directions. In order to determine it accurately, effects of some calculation settings such as simulation cell size, the number of displacement directions and recoil energy increment value for average TDE calculating were analyzed. As a result, we obtained 83 eV as TDE of tungsten. This value is close to the one recommended value by American Society for Testing and Materials (ASTM) which is 90 eV.*

I. INTRODUCTION

In nuclear materials, collision of an incident energetic particle such as high-energy neutron and atom with constituent atoms cause defect formation due to atomic displacement. Formed defects often cause adverse effects on material properties, such as irradiation induced hardening, a shift of the ductile to brittle transition temperature, degradation of the thermal conductivity, etc. In order to control and mitigate adverse radiation effects, it is important to evaluate the types and the number of radiation defects formed during reactor operation.

Threshold displacement energy (TDE) is the minimum recoil energy to displace a lattice atom to an interstitial position in a material, thus to form a stable Frenkel pair. TDE is used in a theoretical model such as NRT model (1) to determine the number of generated defects. Therefore, TDE is regarded as one of the most fundamental quantities in determining the primary state of radiation damage in materials.

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 inconsistency in TDE values reported in previous studies: one study reported around 55 eV (2), and another 123 eV (3. Since this large inconsistency in TDE causes a large uncertainty in the expected number of defects formed in W, it is needed to find an appropriate value for TDE.

Therefore, in the present study, we determine TDE of W using molecular dynamics (MD) method. In MD, TDE is usually defined as an average value of $E_{d,i}$, which is the threshold displacement energy for a specific direction i, over a sufficiently large number of displacement directions. Hereafter, the average value is called $E_{d,avg}$. In order to determine TDE of W as accurate as possible, we analyze effects of MD calculation settings on $E_{d,avg}$ determination. The calculation settings investigated in the present study include a method to search the threshold value in evaluating $E_{d,i}$, simulation cell size, and the number of displacement directions in calculating $E_{d,avg}$. Based on MD simulation results and error analysis results, we discuss causes of the inconsistency in TDE values of W determined in previous studies.

II. DESCRIPTION OF THE ACTUAL WORK

All MD simulations were performed by using the LAMMPS code (5). The interatomic interactions were described with an embedded-atom method (EAM) potential, which was originally parameterized by Derlet et al. (4) and then was revised by Björkas et al. (5) for recoil simulation.

Before initiating a recoil event, the simulation cell was equilibrated with 30 K and 0 Pa, which was used as the initial conditions of recoil simulation. In each recoil event, simulation of around 5 ps was conducted. An adaptive time step was 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 $E_{d,i}$ evaluation in comparison with several other x_{max} and t_{max} settings. We judged whether a defect is formed or not using voronoi analysis implemented in VORONOI package of LAMMPS code.

A recoil MD simulation was initiated 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 was converted to velocity components of PKA when it was given to PKA. Starting with 20 eV, we increased the recoil energy by DE_{step} eV until defects were firstly detected. Then we decreased the recoil energy by 1 eV to reach the minimum energy for defect formation, which is defined as $E_{d,i}$ in the present study.

In order to clarify effects of the calculation settings, we performed the following calculations.

(1) Effect of DE_{step}

DE_{step} is one of calculation setting that affects $E_{d,I}$ evaluated by MD. Figure 1 shows how DE_{step} affects $E_{d,i}$. The blue dots indicate MD simulation results as a function of recoil energy. As depicted in Fig. 1, whether a defect is formed or not does not a simple step function as already indicated in previous studies (6). Therefore, if $E_{d,i}$ is searched with a large DE_{step} value, there is a possibility to miss true $E_{d,i}$ value. In Fig. 1, for example, $E_{d,i}$ is determined as 32 eV with DE_{step} = 1 eV. On the other hand, if DE_{step} = 5 eV is used, the defect is first formed at 45 eV and then $E_{d,i}$ is determined to be 42 eV by decreasing the recoil energy by 1 eV. As seen here, a larger DE_{step} basically overestimate $E_{d,i}$ more strongly.

In order to confirm the effect of DE_{step}, we performed a a set of simulations with chaning DE_{step} from 1 eV to 11 eV. Other calculation settings were fixed as follows: the system size is 8×8×12 supercell; the number of displacement directions is 10000 points.

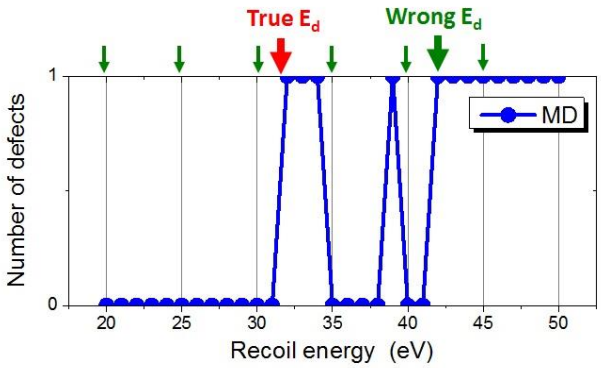

Figure 1. How DE_{step} affects MD simulation result in determination of $E_{d,i}$.

(2) Effect of simulation cell size

In reality, a system is non-periodic and sufficiently large. Instead, in simulations, the periodic boundary conditions are used to model a large system. Under the periodic boundary conditions, a larger simulation cell size is more close to the real system and thus gives a more accurate result. However, as the simulation cell size is enlarged, the computational cost is escalated. Therefore, there is a trade-off between the accuracy and the computational efficiency.

In recoil simulation of bcc metals, <111> recoil easily induces sequential displacement along <111> direction. Therefore, if a cubic simulation cell is utilized in the MD simulation, reentering of atoms from one cell side to another cell side due to the periodic boundary conditions invokes an unphysical collision pattern in the simulation. To avoid this, orthorhombic supercells are usually utilized.

In order to see pure effects of the system size, it is better to fix the cell shape. For this aim, we investigated the system size effect with 2×2×4 (32 atoms), 4×4×6 (192 atoms), 6×6×9 (648 atoms), 8×8×12 (1536 atoms), 10×10×15 (3000 atoms) supercells. In addition, we performed a calculation with 18×16×14 (8064 atoms) supercell. Other calculation settings were fixed as follows: DE_{step} is 6 eV; the number of displacement directions is 10000 points.

(3) Effect of the number of displacement directions

As TDE is defined as $E_{d,avg}$, it is necessary to take a sufficient number of sampling points on displacement directions for averaging. In general, as more displacement directions are involved, the estimate becomes more accurate. In order to estimate this effect, we first generated uniformly distributed points on a unit sphere. Then, we converted coordinates of each point to a displacement direction. By this way, we can prepare a list of uniform displacement directions. When preparing a list, we only took points that satisfies x>y>z and x≥0, y≥0, z≥0 considering symmetry of bcc lattice. The directions prepared in this manner correspond to irreducible crystal directions (ICD). For example, in 10000 uniformly distributed directions, we have around 209 ICDs, which is around 1/48 of the number of original directions.

In order to estimate the effect of the number of displacement directions, we determined $E_{d,avg}$ with 1000, 5000, 10000, and 15000 displacement directions, which correspond to 19, 97, 209 and 329 ICDs, respectively. Other calculation settings were fixed as follows: DE_{step} is 6 eV; the system size is 8×8×12 supercell.

III. RESULTS

(1) Effect of DE_{step}

MD simulation results on $E_{d,avg}$ as a function of DE_{step} are shown in Fig. 2. As DE_{step} increases, $E_{d,avg}$ increases. This trend is reasonable because $E_{d,i}$ search with a larger DE_{step} has a higher possibility to miss the true $E_{d,i}$ value and thus to overestimate $E_{d,i}$ as explained in Fig. 1. Consequently, the average value of $E_{d,i}$ over sampled displacement directions is overestimated with a large DE_{step} value as well. It is reasonable to consider that the true $E_{d,avg}$ is the value at DE_{step} = 0 eV, which can be evaluated by extrapolating simulation results. As a result, the true $E_{d,avg}$ is obtained to be 89 eV, which is the value in 8×8×12 supercell with 10000 displacement directions. It should be noted that the error in $E_{d,avg}$ is twice as large as DE_{step}, approximately. Although the influence of DE_{step} has been hardly investigated in previous studies, its effect should be considered to accurately determine $E_{d,avg}$.

(2) Effect of simulation cell size

MD simulation results on $E_{d,avg}$ with 6 systems of different sizes are shown in Fig. 3. In the smallest system (2×2×4) where only 32 atoms are included, a signifncantly small $E_{d,avb}$ is obtained to be 62.6 eV. This is mainly due to the fact that the system is melt with around 60 eV recoil energy as a heat bath to remove the excess energy from the system was not used in the present study. Once the system is melt, voronoi analysis detects a large number of defects.

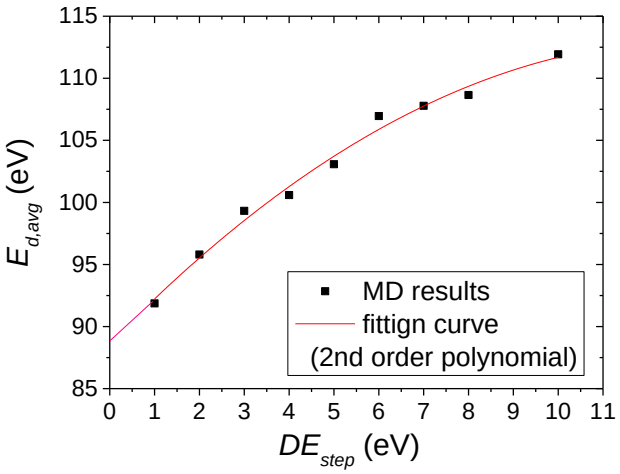

Figure 2. Effect of DE_{step} on determined $E_{d,avg}$

Except for the smallest system, $E_{d,avg}$ monotonically decreases as the system size increases. The monotonic decrease can be explained by considering defect recovery. For a smaller system, a larger temperature increase is induced. As a result, some of created Frenkel pairs are recovered within 5 ps, which causes overestimation of $E_{d,avg}$. $E_{d,avg}$ value converges to around 98 eV as the system size approaches infinity.

In the result of DE_{step} effect (1), we utilized 8×8×12 supercell composed of 1536 atoms. Since $E_{d,avg}$ = 105 eV with 8×8×12 system is larger than the converged value ($E_{d,avg}$ = 98 eV) by around 7 %, we expect a similar error is included in $E_{d,avg}$ = 89 eV determined in (1). Correcting this error, $E_{d,avg}$ is calculated to be 83 eV in a very large system with DE_{step} → 0 eV.

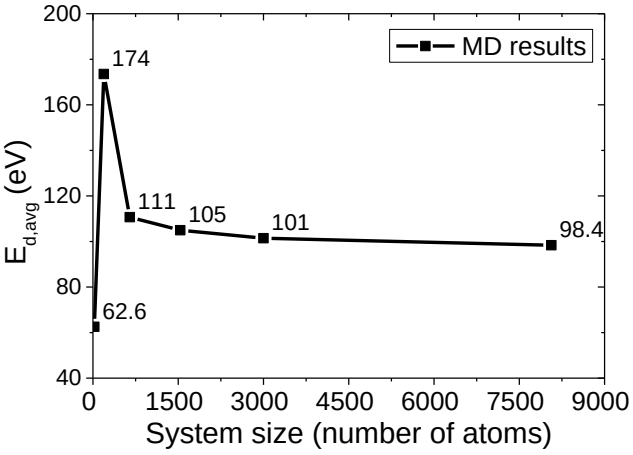

Figure 3. Effect of system size on determined $E_{d,avg}$

(3) Effect of the number of displacement directions

MD simulation results on $E_{d,avg}$ as a function of the number of ICDs are shown in Fig. 4. The dependence on the number of ICDs is relatively weak. Even with the smallest number in the present study, which is 19 ICDs, the calculation result is only different form $E_{d,avg}$ of the largest number, which is 329 ICDs, by around 2%. This could be because even 19 ICDs correspond to 1000 directions and thus are not a small number of sampling points. In the calculations of (1) and (2), we used 209 ICDs. The error expected in the result due to the number of displacement directions would be around 2% at most.

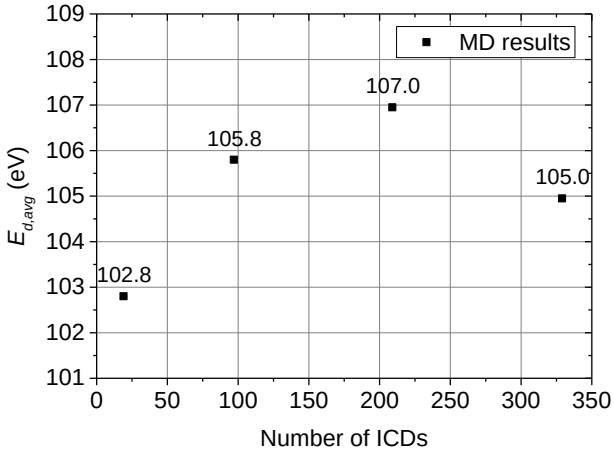

Figure 4. Effect of the number of ICDs on determined $E_{d,avg}$

In summary, the present calculation suggests 83 eV as TDE of W with an error around 2% in it. This estimate was achieved as follows:

a) $E_{d,avg}$ is 89 eV in 8×8×12 system for 209 ICDs, with DE_{step}→ 0.
b) 8×8×12 system size cause overestimation of $E_{d,avg}$ by around 7%. Including this effect brings $E_{d,avg}$ = 83 eV.
c) Determined $E_{d,avg}$ is less dependent on the number of displacement directions. The error expected in the usage of 209 ICDs is less than round 2%.

The obtained $E_{d,avg}$ = 83 eV is comparable with the threshold value recommended by American Society for Testing and Materials (ASTM), which is 90 eV.

In the present study, we did not check potential model dependence of $E_{d,avg}$ evaluation. In a systematic study by Nordlund et al. on the threshold energy evaluated by MD in bcc-Fe, as far as the potential model gives threshold energies for <100>, <110> and <111> directions comparable with experimental values, $E_{d,avg}$ determined by MD is not strongly dependent on the potential model (6). Specifically, 9 $E_{d,avg}$ values determined with 9 different potential models range

from 33.4 eV to 46.3 eV (6), which indicates that the deviation is around ±15%. In the present potential model, the threshold displacement energy for <100> and <111> displacements were calculated to be around 43 eV and 42 eV, respectively. These values are comparable with an experimental result: 42 eV and 41 eV, respectively (7). Relying on this good agreement with experiment, we think the present estimate should be reasonably accurate.

Finally, we discuss the reasons of the inconsistency in previous studies and of agreement and disagreement between the present study and previous studies on the threshold displacement energy of W. Fikar et al. reported the minimum threshold displacement energy over a variety of displacement directions determined by MD (8). They found <100> displacement holds the minimum value, which ranges from 45 eV to 61 eV depending on the potential model (8). Since this result evaluated only the minimum value, not the average value, over many directions, the result corresponds to $E_{d,100}$ in the present study, which is around 43 eV. Thus, the result in ref. (8) is reasonably inconsistent with the present study.

Setyawan et al. reported $E_{d,avg}$ determined by MD in their study focusing on cascade morphology transition in bcc metals (3). $E_{d,avg}$ were obtained to be 122.6 eV with a bond order potential model (9) and 98.0 eV with the same EAM potential model with the present study. As discussed above, this EAM potential model provides good agreement with experimental results on <100> and <111> threshold displacement energies. Thus, we consider 98.0 eV is more reliable than 122.6 eV. The difference between 98.0 eV in ref. (3) and 83 eV in the present study can be ascribed to DE_{step} effect. In ref. (3), DE_{step} = 5 eV was used, which induces around 13 eV error according to Fig. 2. Considering this effect, there is no significant difference between ref. (3) result and the present result.

Mason et al. estimated $E_{d,avg}$ = 55.3 eV using a theoretical equation proposed by Jan and Seeger (10) based on the two experimental displacement energies ($E_{d,100}$ = 42 eV and $E_{d,111}$ = 41 eV (7)) and one guess ($E_{d,110}$ = 70 eV) (2). However, in comparison with the present MD result on direction dependence of $E_{d,i}$, we confirmed that the theoretical model is prone to underestimating $E_{d,avg}$ as it does not consider directions, where relatively larger $E_{d,i}$ emerges, such as <321>. Therefore, the discrepancy between ref. (2) and the present study is ascribed to the error in the theoretical model used in ref. (2).

ACKNOWLEDGEMENT

This research was supported by National Research Foundation (NRF) of Korea under Nuclear Fusion Basic Research program and by BK 21 plus project in Department of Energy Resource Engineering in Seoul National University.

REFERENCES

1. M. J. Norgett, M. T. Robinson, and I. M. Torrens, Nucl. Eng. Des. **33**, 50 (1975).
2. D. R. Mason, X. Yi, M. A. Kirk, and S. L. Dudarev, J. Phys. Condens. Matter **26**, 375701 (2014).
3. W. Setyawan, A. P. Selby, N. Juslin, R. E. Stoller, B. D. Wirth, and R. J. Kurtz, J. Phys. Condens. Matter **27**, 225402 (2015).
4. P. M. Derlet, D. Nguyen-Manh, and S. L. Dudarev, Phys. Rev. B - Condens. Matter Mater. Phys. **76**, 54107 (2007).
5. C. Björkas, K. Nordlund, and S. Dudarev, Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms **267**, 3204 (2009).
6. K. Nordlund, J. Wallenius, and L. Malerba, Nucl. Instruments Methods Phys. Res. Sect. B Beam Interact. with Mater. Atoms **246**, 322 (2006).
7. F. Maury, M. Biget, P. Vajda, A. Lucasson, and P. Lucasson, Radiat. Eff. **38**, 53 (1978).
8. J. Fikar and R. Schäublin, J. Nucl. Mater. **386**–**388**, 97 (2009).
9. N. Juslin, P. Erhart, P. Tr??skelin, J. Nord, K. O. E. Henriksson, K. Nordlund, E. Salonen, and K. Albe, J. Appl. Phys. **98**, (2005).
10. R. V. Jan and A. Seeger, Phys. Status Solidi **3**, 465 (1963).