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Molecular Dynamics Study for the Uniaxial Stress Effect on the Threshold Displacement Energy and Defect Formation Energies in Tungsten

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Abstract - *Defect formation energies and threshold displacement energy (TDE) are important quantities to determine the number of defects formed by irradiation. In this paper, we study effects of uniaxial strain on these quantities in tungsten, which is a candidate material for plasma-facing components in fusion reactors. For tungsten, the most stable defect type for self-interstitial atom is a crowdion. With a uniaxial strain, the formation energy of self-interstitial atom decreases largely, while the vacancy formation energy increases slightly, and the threshold displacement energies of ⟨100⟩, ⟨110⟩, ⟨111⟩ and ⟨321⟩ directions as well as the average of 300 directions decrease concurrently. The results indicate that a compressive stress on tungsten would increase the radiation resistance of tungsten, while a tensile stress would decrease it.*

I. INTRODUCTION

In nuclear materials, the collision of an incident energetic particle such as high-energy neutron and ion 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.

Tungsten (W) is a promising candidate material for plasma-facing components in fusion reactors such as for diverter. In fusion reactors, the diverter will be subjected not only to a 14 MeV neutron flux but also to combined time-varying high thermal fluxes and high mechanical stresses. The combined stresses can affect the formation and properties of radiation-induced defects. In addition, displacement cascades in W would be influenced by such stresses, leading to changes in the predominant structure, concentration and distribution of defects. This influence has been observed in previous studies on alpha-zirconium [2] and copper [3]. Since a large stress is expected to occur in plasma-facing components during fusion reactor operation, the influence of stress field on radiation defect formation in W should be sufficiently understood.

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 [4] 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. In our previous work [5], TDE of bcc-W was determined to be 85 eV with an estimated error of 4.5% for stress-free conditions.

In the present work, the influence of the applied uniaxial strain on the formation energy of simple point defects and TDE are investigated with molecular dynamics (MD) simulations.

II. DESCRIPTION OF THE ACTUAL WORK

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 used the calculation setting that was tested in a previous work [5]. $E_{d.avg}$ value determined in a system under a uniaxial strain is called $\overleftrightarrow{\mathrm{Ed}}_{avg}$.

All MD simulations were performed by using the LAMMPS code [6]. 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. [8] (DB) for recoil simulation.

8×8×12 supercells of BCC-W (containing 1536 W atoms) were used in MD simulations. Before initiating a recoil event, the simulation cell of a non-strained structure was equilibrated with 30 K and 0 Pa under the NPT ensemble. Subsequently, uniaxial deformation strains from −2 % ~ 2.2 % with an interval of 0.6 % were applied to the simulation box. This was made by deforming the simulation cell in the x-direction at a strain rate of 0.002 ps^{-1} under the NVT ensemble. When the simulation cell is stretched along x-direction, the strain perpendicular to the load is kept zero. Thus, the cell vectors along y and z directions are fixed to be the values of the non-strained system throughout the present study.

In each recoil event, a 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. Whether a defect is formed or not was judged by 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 the velocity components of PKA when it was given to PKA. Starting with 10 eV, we increased the recoil energy by 1 eV until a defect is firstly detected. By that the minimum energy for defect formation along a specific displacement direction was determined, which is defined as $E_{d,i}$ in the present study. In the same manner, $\overleftrightarrow{\mathrm{Ed}}_i$ of strained systems were also determined.

The average TDE of 100, 200, 300 and 400 quasi-uniform directions for the strained structures ($\overleftrightarrow{\mathrm{Ed}}_{avg}$) were evaluated. The method to obtain quasi-uniform directions is described in our previous paper work [5]. To find the stable point defect under a constant applied strain field, seven configurations of point defect, i.e., vacancy, ⟨111⟩-crowdion (crowd111), ⟨100⟩-dumbbell (db100), ⟨110⟩-dumbbell (db110), ⟨111⟩-dumbbell (db111), octahedral (oct) interstitial and tetrahedral (tetra) interstitial, were constructed and relaxed by energy minimization methods under the strained systems.

The formation energy of a defect (E_f) can be calculated as

$$\mathrm{E_f = E_{df} - E_{perf}\,\frac{N_{def}}{N_{perf}}} \qquad (1)$$

where E_{df} is the total energy of the system including a defect, E_{perf} is the total energy of a perfect W lattice without defects, N_{def} is the total number of atoms in the defective system, and N_{perf} is the total number of W atoms in the perfect system.

III. RESULTS AND DISCUSSION

(1) Effect of strain on the defect formation energy

The formation energies of the seven point defects with uniaxial strains are shown in Fig. 1. For self-interstitial atoms (SIAs), crowdion and ⟨111⟩ dumbbell formation energy are very close to each other and the most stable configuration is a crowdion, which is the same result found in a stress-free system [9]. For vacancy, a formation energy comparable to experimental result (3.23 eV [10]) was obtained as in Fig. 1

The formation energy of vacancy increases slowly with strain from compression to tension, which is similar to the formation energy of vacancy in copper under uniaxial strain [11]. This is due to the interaction between the local tensile stress induced by the formation of a vacancy and the externally applied stress. If a compression strain is applied, the local tensile stress induced by a vacancy decreases, making vacancy formation easier. A tensile strain causes the opposite effect. At 2.2 % tensile strain, the vacancy formation energy increases by ~10% compared with the value at -2.0% compressive strain. Thus, vacancy formation becomes slightly more difficult if the volume of material increases, while it becomes easier if the volume decreases. Namely, if a W material is put under a tensile stress, the void formation resistance and swelling resistance increase slightly.

For SIAs, the formation energy decreases rapidly with the strain from compression to tension. The trend agrees with the DFT calculation in α-iron [12] under a uniaxial strain. Compared with a vacancy, a SIA induces local compressive stress. Thus, an additional tensile strain can counterbalance the SIA-induced compressive stress, and thereby decreases the interstitial formation energy. Therefore, if W is loaded in a tension stress condition, more interstitials should be formed by collision cascade.

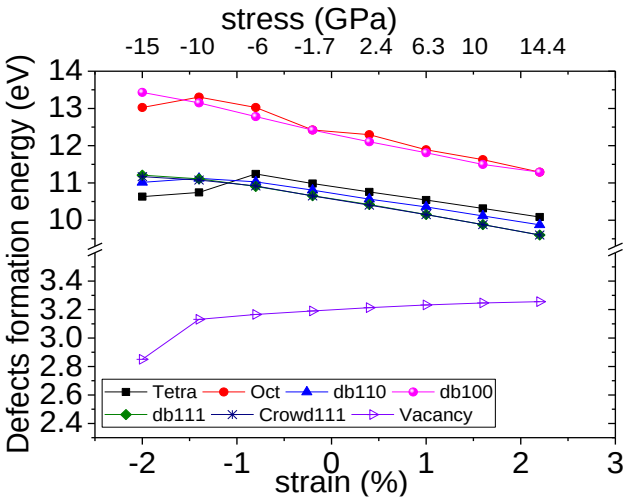

Figure 1. Strain effect on the formation energies of point defects in W. Tetrahedral and octahedral configurations were formed by putting an interstitial atom at (a/4, a/2, 0) and (a/2, a/2, 0), respectively, where a is the lattice constant. 110, 100 and 111 dumbbell configurations were obtained as in ref. [11]. A 111 crowdion was formed by inserting an extra atom halfway between the nearest neighbors.

(2) Effect of strain on TDE of specific displacement directions

Four displacement directions, i.e., ⟨100⟩, ⟨110⟩, ⟨111⟩ and ⟨321⟩, were simulated. The threshold energy for a specific direction i under an applied stress is expressed as $\overleftrightarrow{\mathrm{Ed}}_i$ in this paper. The calculated results for each direction under applied strains are shown as a function of stress in Fig. 2. The $\overleftrightarrow{\mathrm{Ed}}_i$ values slowly change for both of ⟨111⟩ and ⟨100⟩. On the other hand, for ⟨110⟩ and ⟨321⟩ recoils, the $\overleftrightarrow{\mathrm{Ed}}_i$ values change largely and non-smoothly.

Since the crowdion is the most stable self-interstitial configuration in W, the majority of defects produced in a displacement cascade should be crowdions and vacancies. Hence, it is reasonable to define the formation energy of Frenkel pair as the summation of the crowdion and vacancy formation energies. In Fig. 2, the estimated Frenkel pair formation energy is compared with $\overleftrightarrow{\mathrm{Ed}}_i$.

From Fig. 1 and 2, we can see clearly that the decrease of SIA formation energy with strain is faster than the increase

of vacancy formation energy. As a result, the formation energy of the Frenkel pair decreases with strain from compression to tension, resulting in a decrease of $\overleftrightarrow{Ed}_i$ values.

The minimum threshold displacement energy is achieved along the ⟨111⟩ direction for non-strained system [5]. This fact is not changed by the uniaxial stresses.

Compared with the formation energies of point defects, the effect of applied strain on TDE is similar to its effect on SIAs. As $E_{d,i}$ is the minimum energy to produce a stable Frenkel pair, when strain is applied, the formation energy of a Frenkel pair is changed. When W is loaded in compression, more energy is needed to create a stable Frenkel pair, which may lead to the decrease of defect production. The investigation of iron by Beeler et al. [1] indicates that the strain presents a similar effect on the formation energy of the Frenkel pair and defect production.

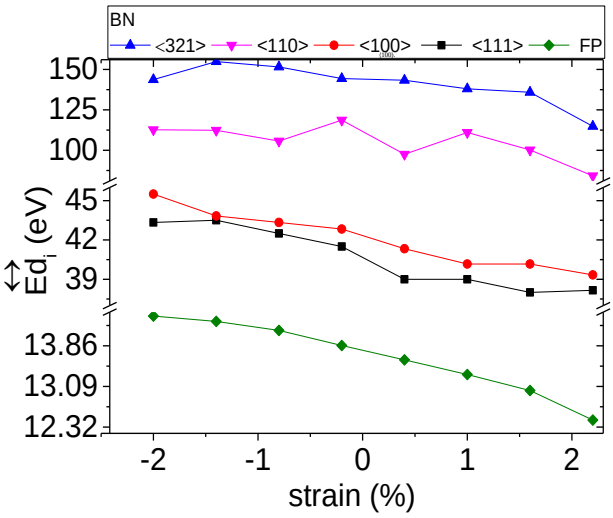

Figure 2. Strain dependence of the threshold displacement energies of ⟨100⟩, ⟨111⟩, ⟨110⟩, and ⟨321⟩ directions

(3) Effect of strain on average TDE

In determining $\overleftrightarrow{Ed}_{avg}$, 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 determine the number of sampling directions for accurate estimation of $\overleftrightarrow{Ed}_{avg}$, we determined $\overleftrightarrow{Ed}_{avg}$ with 100, 200, 300 and 400 quasi-uniformly distributed directions.

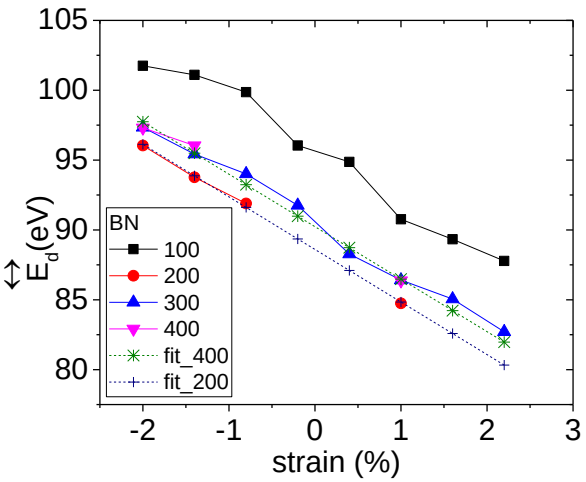

Figure 3. Effect of the number sampling directions on determined $\overleftrightarrow{Ed}_{avg}$

The profiles obtained with 300 directions reasonably agree with that obtained with 400, while that obtained with 100 does not. This means that 100 does not appropriately sample the probability density function of $\overleftrightarrow{Ed}_i$. We considered that 300 sampling directions are enough to determine $\overleftrightarrow{Ed}_{avg}$ within an error of around 1 eV.

As the same with TDE for specific directions ($\overleftrightarrow{Ed}_i$) shown in Fig. 2, $\overleftrightarrow{Ed}_{avg}$ decreases as the strain increases. As discussed in Section II. (2), the decrease can be related to the decrease of Frankel formation energy. Specifically, the Frenkel formation energy and $\overleftrightarrow{Ed}_{avg}$ decrease by 16% and 18%, respectively, from -2.0% strain to 2.2% strain.

In summary, the effect of uniaxial strain on defect formation energy and the threshold displacement energy of BCC W has been studied with the MD simulation method. With the uniaxial strain ranging from 2 % compression to 2.2 % tension, the formation energies of self-interstitials decrease rapidly, while the vacancy formation energy increases slowly, and the threshold displacement energies of ⟨100⟩, ⟨110⟩, ⟨111⟩ and ⟨321⟩ directions as well as the average TDE decrease concurrently. Independent of strains, the most stable SIA-type point defect is a crowdion and the minimum $E_{d,i}$ is achieved along ⟨111⟩ direction. The present MD simulation results indicate that a compressive stress on W would increase the radiation resistance of W, while a tensile stress would decrease it.

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