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Analysis of Calculation Errors in Evaluation of Threshold Displacement Energy by Molecular Dynamics in BCC-Fe

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1. Introduction

In nuclear fission/fusion materials, defects are created by irradiation of energetic particles during the reactor operation. The accumulation of radiation defects causes adverse changes in mechanical and thermal properties of the materials such as hardening or decrease of thermal conductivity. As the accumulation amount is determined by a balance between the formation and the annihilation of radiation defects, it is important to know how many defects are formed by the irradiation.

In the evaluation of the number of formed radiation defects, the most fundamental property is the threshold displacement energy (TDE). It is defined as the minimum kinetic energy to displace an atom from its lattice site to a defect site. In other words, TDE corresponds to the minimum energy to form a pair of vacancy and self-interstitial-atom (SIA), namely a Frenkel pair. The amount of radiation defects is calculated with TDE in the Kinchin-Pease model and the Norgett-Robinson-Torrens (NRT) model [1]. Although some faults are known in these classical models [2], they are still widely used to analyze the radiation effect in materials and then to compare the effects of different radiation conditions.

The TDE has been determined both by experiments and by simulations in various materials. In experiments there are several methods to determine TDE, such as using the resistivity measurement of electron-irradiated samples and using a high-voltage electron microscope (HVEM). However, to determine a TDE by experiment, both defect creation and defect detection need to be performed precisely, which is a formidable task.

In simulations, molecular dynamics (MD) has been widely utilized. It has advantages over experiments on its low cost and short time as well as on its accessibility to the dynamics of atoms in an event of radiation, which enable us to understand details of radiation damage processes. However, there are several subjects that need to be solved to use the MD for this purpose. For example, Nordlund *et al* [3] showed that TDE calculated by MD largely depends on potential models. 11 tested potential models gave different TDE values, which ranged from 22.0 eV to 53.5 eV.

In the present study, we investigate the accuracy and precision of TDE determined by MD. We chose bcc-Fe as the target system, which has been widely studied as a model system of ferritic steels used for reactor pressure vessels in nuclear fission reactors and blanket structures in nuclear fusion reactors. Therefore, radiation damage in iron is a passionate concern for both nuclear fission and fusion researchers.

2. Methods

MD simulations were performed using the Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS) code. An embedded atom method (EAM) potential parameterized by Björkas *et al* for iron was used [4].

The simulation cells used in the present study were N×N×1.5N orthorhombic systems (N=4, 6, 8), whose sizes range from 192 to 1536 atoms. The initial temperatures were set to be 30 K or 0 K. For 30 K simulations, prior to a recoil simulation, the system was equilibrated under a canonical (NVT) ensemble implemented with a Nose-Hoover thermostat at 30 K. In the 0 K simulations, a perfect crystal was used as the initial structure.

A recoil simulation was started by giving an excess kinetic energy to a primary-knock-on-atom (PKA). The recoil MD simulation was performed under the micro-canonical (NVE) ensemble. During the recoil simulation, two characteristic phases basically appear, namely collision phase and relaxation phase. In the collision phase, the PKA energy is distributed by few kinetic collisions among atoms around PKA. Some of the collided atoms are displaced from lattice sites. In the relaxation phase, the kinetic and potential energies reach steady states in the system. Before reaching such steady states, most displaced atoms return to lattice site, and only some stable defects may remain. We confirmed that 4 ps after the start of recoil simulation was an appropriate timing for judging whether a stable defect is created or not. The defect creation was judged by the Wigner-Seitz cell analysis using Voro++ package [5].

In the present study, we define the TDE as the minimum energy that stable defect is detected. If no stable defects remained at the end of a recoil simulation, a next recoil simulation was performed with a higher PKA energy. This was repeated until a stable defect was detected. The increment of a PKA energy was set from 1 to 4 eV. After finding the energy to form a stable defect, the PKA energy was decreased by 1 eV until the energy that causes no defect was detected. In this manner, a threshold energy was determined

The effective value of TDE (E_d), which is an input for the Kinchin-Pease model and the Norgett-Robinson-Torrens (NRT) model, was determined as the average value of TDEs over various directions ($N_{direction}$).

$$E_d = \frac{1}{N_{direction}} \sum_i E_{d,i} \qquad (1)$$

where $E_{d,i}$ is TDE of i-th direction.

In the present study, we investigated four points related to the precision and accuracy of TDE calculated by MD: effects of (i) direction dependence, (ii) energy incremental interval, (iii) thermal vibration, and (iv) potential model on the TDE calculation.

Firstly, we checked how TDE depends on the displacement direction, and how many $N_{direction}$ is needed to sufficiently represent all directions. The number of directions must be large enough and the sampled direction must be uniform enough. For this purpose, a set of almost uniform directions were constructed by locating point charges and minimizing the total energy of point charges step by step [6].

Secondly, we evaluated how the energy increment interval changes E_d. If a larger value is used for the interval, there is a larger possibility to miss a true threshold value.

Thirdly, we investigated how the thermal vibration affects TDE. If an MD recoil simulation is performed at a finite temperature, like 30 K, the structure when a recoil event is initiated varies by thermal vibration. Thus, to accurately determine $E_{d,i}$ and E_d, we need to perform several simulations and then to take an average of these simulation results.

Fourthly, we have been investigating how the potential model used in the MD simulations affects TDE. Indeed, this is one of the largest error source when the MD simulation is applied to the evaluation of the TDE.

3. Results and discussion

3.1. Effect of displacement direction

Fig. 1 shows direction dependence of $E_{d,i}$ and the standard deviation (σ_i) that was obtained from simulations of 210 directions, 1000 timings for each direction, and 4 eV energy increment interval. A significant directional anisotropy is observed.

$E_{d,i}$ takes a local minimum at the <100> direction and a local maximum at the <111> direction. Indeed, the <100> direction corresponds to the global minimum. Around each of these two directions, similar $E_{d,i}$ values are obtained. In addition, the standard deviations are relatively small for around these directions.

The highest $E_{d,i}$ appears around <321> direction. The σ_i is also high in this region as shown in Fig. 1(b). In Fig. 1(a), the interval of contour lines are narrow for around this direction, which means that $E_{d,i}$ can easily change if a displacement direction is slightly altered.

Due to these large directional anisotropy, we need to make simulations of many directions to appropriately determine E_d by Eq. (1). We decided to perform simulations with $N_{direction}$ = 210. This setting causes an error up to around 1 eV in E_d, which is sufficient for the purpose of the present study.

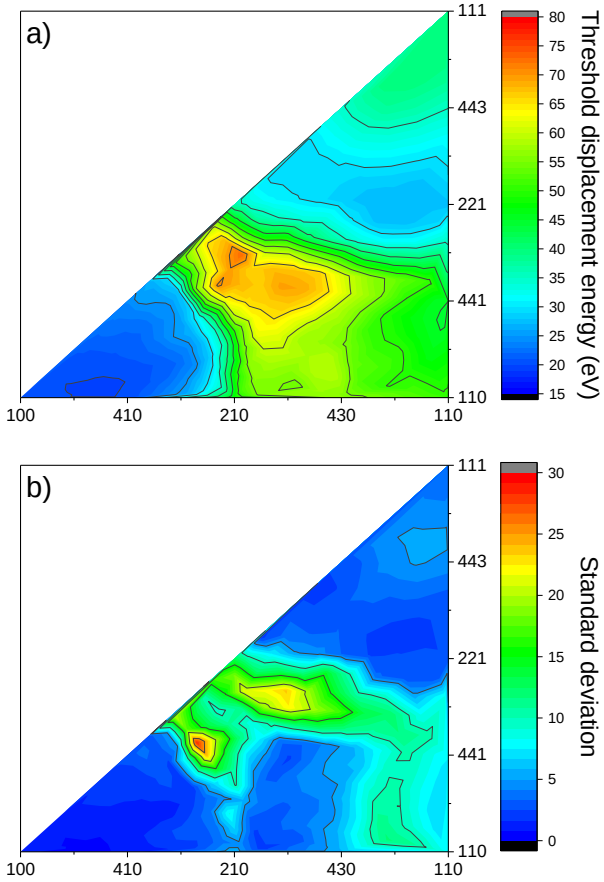

Figure 1. Directional dependence of $\boldsymbol{E_{d,i}}$ and $\boldsymbol{\sigma_i}$.

3.2. Effect of energy increment interval

Fig. 2 plots calculated E_d as a function of energy increment interval. The calculations were conducted in simulation cells of three different sizes. The energy increment interval gives almost the same effect to TDE between the 6×6×9 and 8×8×12 systems. This result indicates that the system size effect is negligible for cells larger than 6×6×9. The 4×4×6 system seems not to be appropriate to determine TDE in bcc Fe.

The error due to the energy increment interval is expressed as follows:

$$\Delta_{ei} = 0.82 E_{in}. \qquad (2)$$

The true value, which is used as the reference value in the error estimation, is considered to be 39.2 eV. The true value was obtained by extrapolating the fitting lines for the 6×6×9 and 8×8×12 systems to E_{in} = 0 eV because the error caused by the energy increment interval is expected to be 0 when the interval is set to be 0 eV.

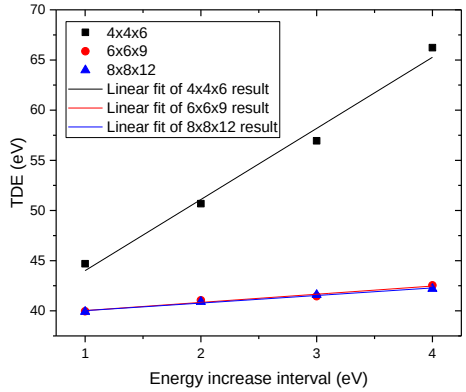

Figure 2. $\boldsymbol{E_d}$ as a function of energy increment interval in 4×4×6, 6×6×9, 8×8×12 unit cells.

3.3. Effect of thermal vibration

Fig. 3 shows E_d values calculated by changing the timing to introduce a PKA energy in the 30 K simulation. The tested timings ranged from 0 fs to 999 fs. The minimum and maximum values of E_d were 38.38 eV and 44.64 eV, respectively. The results of close timings seem to be correlated with each other in Fig. 3.

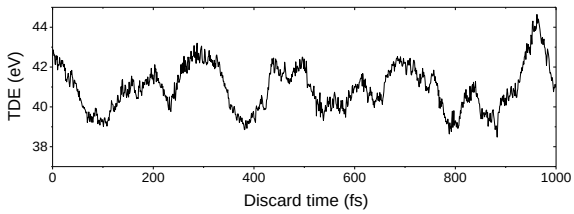

Figure 3. Vibration effect on $\boldsymbol{E_d}$. $\boldsymbol{E_d}$ is averaged over 210 directions with each timing.

In order to analyze the characteristic frequency in the profile presented in Fig. 3, the data was processed by the fast Fourier transform (FFT). The transform showed that the main component is located at $0.004\ fs^{-1}$ (=$4.0 \times 10^{12}\ Hz$). This frequency is similar to a typical frequency of atoms in the bcc-Fe lattice, which is calculated as [7]:

$$f_{Fe} = \frac{1}{2\pi}\sqrt{\frac{aE}{m}} = \frac{1}{2\pi}\sqrt{\frac{48.12\ N/m}{9.27\times10^{-26}\ kg}} = 3.6 \times 10^{12}\ Hz, \quad (3)$$

where a is the interatomic spacing (2.28 Å), E is Young's modulus (211.4 GPa), and m is the atomic mass of Fe (9.27×10^{-26} kg/atom).

As the timing has a large effect on the simulation results, we investigated an appropriate sampling criterion for the collision event timings. First, we reconstructed the E_d profile of Fig. 2 based on the FFT result as follows:

$$y_{recon}(t) = \sum_{k=0}^{N_{wave}} A_k \cos(2\pi(f_k t) + \varphi_k), \quad (4)$$

where N_{wave} is the number of cosine waves, A_k is the k-th cosine wave's amplitude, f_k is the k-th cosine wave's frequency, and φ_k is the k-th cosine wave's phase angle. The difference between the simulation data given in Fig. 3 and the reconstructed profile given by Eq. (4) was almost negligible. Therefore, we used the analytical expression by Eq. (4) in the analysis of the sampling criterion. With Eq. (4), the total uncertainty (σ_{tot}) with sampling interval Δ and the number of samples $N_{sampling}$ is expressed as

$$\sigma_{tot}(\Delta, N_{sampling}) = \sqrt{\sum_{k=0}^{N_{spectrum}} [\sigma_k(\Delta, N_{sampling})]^2}, \quad (5)$$

where σ_k is the uncertainty of the k-th cosine wave in Eq. (4).

Fig. 4 shows calculated σ_{tot} as a function of Δ and $N_{sampling}$. Basically, σ_{tot} decreases as '$\Delta \times N_{sampling}$' value increases. The decrease is affected not only by $N_{sampling}$ but also by Δ. This result suggests that E_d values are time-correlated data, as expected from Fig. 3.

Fig. 5 shows σ_{tot} as a function of Δ with fixing $N_{sampling}$ to be 20 and 40. This result indicates that the time-correlation is largely reduced by around 25 intervals. Using shorter interval than 25 fs would be ineffective to reduce the thermal vibration effect.

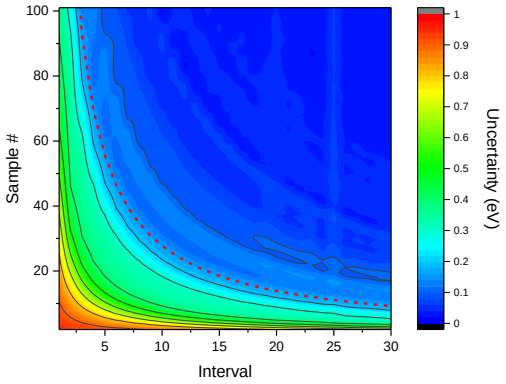

Figure 4. The total uncertainty of $\boldsymbol{E_d}$ by sampling conditions. The red dash line indicates '$\boldsymbol{\Delta \times N_{sampling} = f_{Fe}}$'.

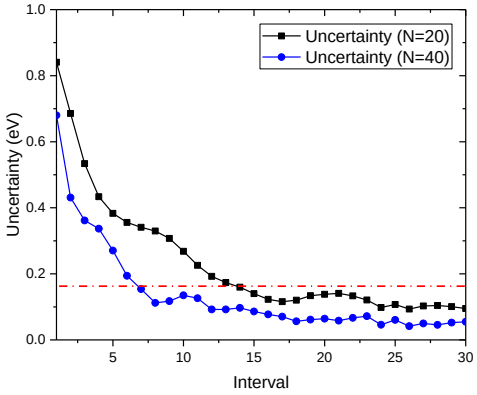

Figure 5. Variation in the uncertainty of E_d as a function of sampling interval with fixing $N_{sampling} = 20$ & 40.

3.4. Effect of potential model

In a previous experimental study by F. Maury *et al* [8], $E_{d,<111>}$ was obtained as 20 eV. However, $E_{d,<111>}$ in this study is much higher than that of previous study. The difference between experiment and simulation may come from the inaccuracy of the present potential model.

To check this possibility, we performed MD simulations with several different potential models. With almost all potential models, $E_{d,<100>}$ was a local minimum and showed a relatively small (or the smallest) value in all the tested displacement directions. On the other hand, $E_{d,<111>}$ became a local maximum with some potential models including the present model, while it became a local minimum with other potential models. We will further analyze this result and will additionally present it in the meeting.

4. Conclusion

In the present study, we have investigated four possible error sources in the evaluation of TDE by MD. Firstly, we confirmed a strong directional anisotropy of $E_{d,i}$ and σ_i. The largest $E_{d,i}$ appeared around <321> direction, where the σ_i value was also high. 210 sampling direction was sufficient to reduce the error in E_d below around 1 eV. Secondly, we analyzed the error induced by the energy increment interval, which was expressed as a linear function of energy increment interval as given in Eq. (2). Thirdly, the thermal vibration effect of E_d was studied. The maximum and minimum E_d were obtained to be 44.64 eV and 38.38 eV in the calculations for 1000 timings of 1 fs intervals. We analyzed the time-correlation in E_d values and suggested a criterion to reduce the correlation. Lastly, we observed that $E_{d,<111>}$ was largely dependent on the potential model. This will be further studied and additionally presented in the meeting.

5. Acknowledgement

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