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First-principles and classical molecular dynamics study of threshold displacement energy in beryllium

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ABSTRACT

Beryllium selected as a neutron multiplier material for the tritium breeding blanket of fusion reactor should withstand high doses of fast neutron irradiation. The damage produced by irradiation is usually evaluated assuming that the number of atomic displacements to the threshold displacement energy, E_d, which is considered as an intrinsic material parameter.

In this work the value of E_d for hcp beryllium is estimated simultaneously from classical and first-principles molecular dynamics simulations. Quite similar quantitative pictures of defect production are observed in both simulation types, though the predicted displacement threshold values seem to be approximately two times higher in the first-principles approach. We expect that, after more detailed first-principles investigations, this approach can be used for scaling the damage prediction predictions by classical molecular dynamics, opening a way for more consistent calculations of displacement damage in materials.

1. Introduction

Secure functioning of beryllium in nuclear facilities requires deep understanding of the mechanisms of displacement damage generation at the atomic scale. Such information is hard to obtain experimentally and, therefore, the efficiency of radiation damage production is commonly deduced from theoretical considerations or from atomistic simulations. For example, there exists a theory-based Kinchin-Pease equation for the estimation of the number of atomic displacements, $\nu(E)$, produced by an ion with the initial kinetic energy E, so that the total rate of defect production can be estimated for any known spectrum of primary knocked-on atoms (PKAs). This equation is written down as

$$\nu = \kappa \frac{E}{2E_d} \tag{1}$$

where κ is the numerical factor that depended on the particular approximation (e.g. $\kappa = 0.8$ for the commonly used standard Norgett-Robinson-Torrens formulation [1]) and the relation $E > 2E_d$ is assumed. The equation has the only free parameter, E_d, that is usually interpreted as the minimum energy to be transferred to a target atom in order to irreversibly displace it out of its lattice position, creating a stable Frenkel pair.

In practical calculations one usually needs not the numbers of displacements as such, but the numbers of interstitials and vacancies that remain in the matrix after the passage of a PKA. The most common way to estimate the damage rate in terms of point defect production is the use of binary collision (BC) codes (such as SRIM [2]) or molecular dynamics (MD) simulations. Remarkably, the linear or near-linear dependence of the number of created point defects on the primary ion kinetic energy is a common feature of both BC and MD simulations, but the selection of an appropriate value of E_d is important for realistic estimates via BC, where the displacement threshold energy is explicitly used. Hence, the knowledge of realistic E_d is a prerequisite for the reliable estimation of point defect production rate. Unfortunately, the default values of E_d implemented in BC codes (e.g. SRIM) can be quite different from the predictions of detailed atomistic simulations.

A straightforward way to determine the threshold displacement energy is the use of dynamic atomistic calculations, which has already been applied to a number of materials, including Fe [3,4], Si [5], W and V [6], FeCr alloy [7], etc. Strictly speaking, in crystalline materials this lowest energy is sensitive to the direction of the initial recoil, so an additional averaging over recoil flight directions are usually necessary, which is a non-trivial task [3]. First of all, contrary to the intuitive assumption invoked in the concept of

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threshold displacement energy, the probability of defect creation is not necessarily a step function of energy. Second, the selection of weight function for different crystallographic directions is not obvious, especially when one is interested in the simulation of electron scattering experiments on single crystals. For hcp metals, the measurements of direction-dependent displacement thresholds [8,9] constitute a noteworthy example.

The main problem of atomistic techniques for the determination of E_d is the relevant description of atomic interaction dynamics in the collisions caused by the primary recoil. Very accurate interaction description by first-principles density functional theory (DFT) is only rarely used for full-scale simulations [4,5] due to strong computational limitations. More common is the use of semi-empirical interaction potentials that allow easy accumulation of large statistics on direction-dependent displacement thresholds. However, due to an uncertain reliability of such potentials for the description of strong scattering events, the threshold energy predictions are usually sensitive to the particular potential [3,6].

In this work we estimate the value of the threshold displacement energy in Be, using a combination of semi-empirical and first-principles MD simulations. Here, the semi-empirical simulations are used in order to scan the dynamics of damage production on a sufficiently dense net of primary recoil directions, whereas DFT-based molecular dynamics (MD DFT) is used to get more reliable threshold values in the 'low threshold' directions that provide the main contribution to E_d as well as to understand main mechanisms of defect formation.

2. Simulation methods

MD DFT simulations were performed using the MD technique implemented in the plane-wave ab initio code VASP. It is easy to verify that with the beryllium recoil kinetic energies used here (well below 100 eV), the adiabatic approximation implicitly involved in the atomic force calculations holds true. The generalized gradient approximation with the exchange-correlation functional of Perdew–Burke–Ernzerhof (PBE) [10] was applied. To represent the core electrons of beryllium atoms, we used the projector augmented wave (PAW) method [11,12]. The kinetic energy cutoff of 250 eV was applied in all MD calculations. Sampling of the Brillouin zone was performed with the use of a gamma-centered Monkhorst–Pack k-space grid [13].

Simulation cells for DFT calculations contained typically 128 Be atoms in orthorhombic $4 \times 4 \times 4$ unit cells, but for some special recoil momentum directions alternative supercell shapes, elongated in the direction of the PKA movement were used (e.g. $16 \times 2 \times 2$ unit cells for recoils in the $\langle 11\bar{2}0 \rangle$ direction). The $7 \times 7 \times 11$ k-point grid density was used for the basic simulation cell, but it was modified appropriately in elongated supercells.

Each simulation run was launched by assigning to an atom in a static ideal crystal lattice (corresponding to 0 K) a momentum in a pre-selected direction, corresponding to the desired energy. During the MD run, NVE ensemble was used. We used varying time steps (in the range of 0.1–1.0 fs) depending on the PKA energy, in order to ensure that no atom travels more than 0.1 Å per timestep. The typical simulation run was 200–300 steps covering up to 300 fs. In most cases this duration was sufficient to decide whether the produced defects survive or annihilate. In case of doubts, an additional static relaxation of final configuration was performed with a more rigorous parameter set, in order to ensure the reliability of results.

The conventional MD was performed using the LAMMPS code [14] and the ABOP interatomic potential from Ref. [15]. Thirty primary recoils were launched simultaneously in a 30,000 atom orthorhombic simulation box consisting of $5 \times 6 \times 1$ subblocks, with each subblock containing $10 \times 10 \times 5$ parallelepiped unit cells. The periodic boundary conditions in all three dimensions were applied. The sizes of subblocks were selected such that no collision sequence would intervene with a neighbour sub-block.

The collision events were launched by simultaneously assigning to the central atoms of each subblock increasing velocities, directed in the same way and corresponding to recoil kinetic energies from 1 to 30 eV, with a step of 1 eV. Thus, the damage events for each direction of recoil momentum, specified by the azimuthal angle θ and the polar angle φ, were calculated within one MD run. Having in mind the symmetry considerations, θ was varied from 0 (along the c-axis) to 90° (in the basal plane) with the step of 5°. For each azimuthal angle, φ was varied from 0 to 60° between two neighboring $\langle 1\bar{1}00 \rangle$ directions. The number of covered polar angle points depends on θ, following the $\sin\theta$ distribution and changing from one at $\theta = 0°$ to 61 at $\theta = 90°$. The used MD time step was equal to 1 fs and the whole run length was typically 2 ps. In order to eliminate possible remaining unstable interstitial-vacancy pairs, the energy minimization was performed on the final configuration of each run. The identification of interstitials and vacancies in the final configurations was done using the technique suggested in Ref. [16].

3. Results and discussion

3.1. Empirical potential based molecular dynamics

The conventional MD simulations were applied to perform a systematic study of the displacement threshold dependence, using a relatively dense grid of possible momentum directions, as described in Section 2.

A remarkable feature of the obtained results is the fact that in many directions the probability to create a Frenkel pair is not a step function of the primary recoil energy. This is illustrated in Fig. 1 that shows the dependence of the number of created Frenkel pairs as a function of the recoil energy for one of recoil momentum orientations. One can observe several gaps in the defect production probability after the first defect pair is created. This kind of behavior, which was observed earlier also in Ref. [3], demonstrates the sensitivity of defect production at relatively low recoil kinetic energy to fine details of atomic collision kinematics.

With discontinuous probability of defect production, it becomes uncertain, which energy should be considered as the threshold one. In the processing of results, we prefer to eliminate 'stand-alone' events at low recoil energies, when determining the threshold for a particular direction. On the other hand, occasional narrow gaps in the otherwise continuous probability at higher energies are

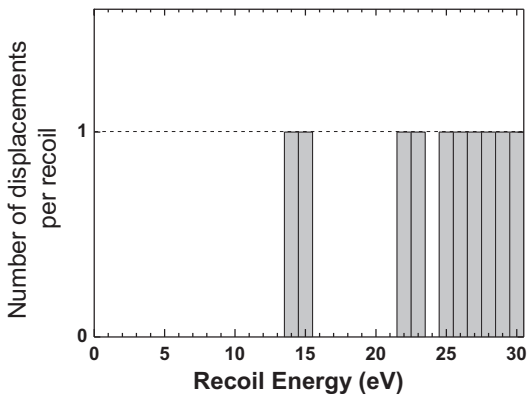

Fig. 1. The number of interstitials produced by primary recoils of different energies and the momentum directed at the polar angle $\varphi = 8°$ in the basal plane.