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Defect-induced change of temperature-dependent elastic constants in BCC iron

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ABSTRACT

The effects of radiation-induced defects (randomly distributed vacancies, voids, and interstitial dislocation loops) on temperature-dependent elastic constants, C_{11}, C_{12}, and C_{44} in BCC iron, are studied with molecular dynamics method. The elastic constants are found to decrease with increasing temperatures for all cases containing different defects. The presence of vacancies, voids, or interstitial loops further decreases the elastic constants. For a given number of point defects, the randomly distributed vacancies show the strongest effect compared to voids or interstitial loops. All these results are expected to provide useful information to combine with experimental results for further understanding of radiation damage.

1. Introduction

Formation of supersaturated point defects (self-interstitial atoms and vacancies) through the interaction between high-energy particles and atoms in materials has been recognized to affect the properties of materials in nuclear reactors, e.g. embrittlement, hardening, swelling, irradiation creep, and so on, resulting in radiation damage [1–11]. In experiments, transmission electron microscope (TEM) is usually used to characterize the evolution of microstructure [1,4,6,8–10]. The stress versus strain (σ vs. ε) curve [12], charpy impact test [13], nanoindentation [14], and irradiation creep test [8] before and after radiation are the main measurements to estimate the radiation-induced mechanical property change. However, these experiments are all destructive methods. Historically, measurement of the change of elastic constants has been used to interpret radiation damage [15,16], which is considered to provide more information than the measurement of electrical resistivity change [15]. Recently, measurement of elastic modulus with surface acoustic wave spectroscopy has been used again in combination with density functional theory (DFT) calculations in W-Re alloy [17] to understand swelling. It should be noted that this experimental method is non-destructive and may be extensively used in future by combining with simulation results. However, the relationship between radiation damage and elastic modulus change (EMC) should be established in advance for its further application.

Because of the structural complexity induced by radiation damage (e.g. supersaturated interstitials, vacancies, voids, and dislocation loops) and the effect of dislocation pinning [18], simulations are needed to complement the experiments to understand the effect of each defect type on EMC. With the development of simulation methods, the properties of radiation-induced defects have been extensively studied theoretically. For examples, the formation and migration energies of defects and the interaction energies between defects have been calculated for iron-based materials [19–24]. In BCC iron, the formation energy of a vacancy (an interstitial) is 1.71 (3.59) eV calculated with molecular dynamics (MD) [25], and 2.07 (3.64) eV calculated with DFT [26], while the migration energy of a vacancy is 0.63 eV with MD [25] and 0.67 eV with DFT [26], and the migration energy of an interstitial is 0.34 eV with DFT [26]. The formation of interstitial dislocation loops is also reported to affect the gliding of dislocations [22,23]. Thus, the shifts of mechanical properties (e.g. radiation hardening and creep), are expected to be affected by the

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distributions of defect density and size, especially the extended defects (voids and dislocation loops). However, the possible relationship between EMC and the density and size of these defects has not been fully studied. Thus, it is necessary and important to establish such relationship in order to use the surface acoustic wave spectroscopy and other similar experimental methods to provide more understanding of radiation damage. In this paper, the effects of vacancies, voids, and interstitial dislocation loops on elastic constant change (ECC) at different temperatures in BCC iron are studied.

2. Simulation method

There are mainly three methods to calculate the elastic constants of a system containing various defects: (1) with the method developed by Daw and Baskes [27]; (2) through the calculation of strain energy for various strained states as suggested by Ackland [28], and (3) by the method developed by Ray et al. [29]. In the first method, only the contribution from static relaxation is considered while in the second method, the temperature fluctuation is included. We follow the third method which will be described below. In addition, due to the complexity of defect type, size, and density, we employed MD method to explore the radiation-induced ECC. The empirical Ackland-2004 Fe potential [25] is used in the present work. Although there are other empirical potentials for iron [30–33], Ackland-2004 is selected because it can reproduce the experimental data of the dependence of elastic constants on temperature for perfect BCC iron better than other potentials. These results are shown in supplementary materials by comparing with the results from Ref. [34]. The computational box is cubic with the X, Y, and Z axes oriented along the [100], [010], and [001] directions, respectively. Periodic boundary conditions are applied along these three directions. There are 20 unit cells along each direction, i.e. 16,000 atoms in a perfect matrix. The defects are then introduced into the matrix by inserting or removing a given number of atoms accordingly. Although DFT or first-principle MD (FPMD) can provide data for small system, it is computationally impractical for a relatively large system, such as 16,000 atoms, needed to avoid the interaction of a defect with its periodic images. The elastic constant tensor, C_{ijkl}, calculated with the method developed by Ray et al. [29] is given as:

$$C_{ijkl} = -\frac{V_0}{k_B T}\delta(P_{ij}P_{kl}) + \frac{2Nk_BT}{V_0}\left(\delta_{il}\delta_{jk} + \delta_{ik}\delta_{jl}\right) + \frac{1}{V_0}\left\langle \sum_{b>a} f\left((r_{ab})x_{abi}x_{abj}x_{abk}x_{abl}\right)\right\rangle_{av} \quad (1)$$

where P_{ij} is the stress component of σ calculated by Parrinello-Rahman method [35] which has included the contributions from momentum and force of N particles in volume V_0. f denotes $r^{-2}(u'' - r^{-1}u')$. u is the potential energy and the last term in RHS of equation (1) is then equivalent to equation (8) defined in Ref. [27] by Daw and Baskes. $\bar{x}_{ab}$ is the vector connecting particles a and b with length r_{ab}. k_B is Boltzman constant and T is temperature. δ is defined with following equation: $\delta(\alpha,\beta) = \langle\alpha\beta\rangle_{av} - \langle\alpha\rangle_{av}\langle\beta\rangle_{av}$. δ_{ij} is Kronecker delta. It should be noted that the temperature effect has also been included in this formula, that is, at each temperature, the elastic constants calculated with this method are isothermal properties, which is consistent with experimental measurement [17]. Because of the limitation of MD method, the quantum statistical effect and zero-point energy are not included. However, based on previous studies [36], these effects would not affect the conclusions obtained in this study. In this equation, the elastic constant is a fourth-order tensor and there are three independent elastic constants in cubic crystals: $C_{11} = C_{1111}$, $C_{12} = C_{1122}$ and $C_{44} = C_{2323}$.

In the present work, the following defects are investigated: (1) vacancies at various concentrations, (2) a void with different sizes, and (3) 1/2 ⟨111⟩ and ⟨100⟩ interstitial loops with different radii. For the first case, the vacancies are introduced into the matrix randomly with a separation distance more than 7 Å between vacancies. In the following, the Vac_R is used to denote such situation. The computational boxes with the number of vacancies from 1 to 320 are calculated. Since the interstitials prefer to form dislocation loops, the effect of interstitials with different concentration is not considered here but rather the effect of interstitial prismatic loops with different radii is simulated. The number of interstitials in a loop is set from 7 to 151 for the 1/2 ⟨111⟩ loop and from 37 to 161 for the ⟨100⟩ loop. For systems with a void, the void is assumed to be a single spherical shape and denoted as Void_S. The number of vacancies in a void embedded in the matrix is varied from 15 to 331. After the defects are introduced, the box is firstly relaxed with molecular static (MS) method. The formation energy of these defects are calculated with the following equation:

$$E^f(defect) = E_{tot}(defect) - (N \pm m) \times E_{perfect} \quad (2)$$

where $E_{tot}(defect)$ is the total energy of the system which contains m vacancies or interstitials. N is the number of atoms in a perfect matrix. Thus, $N \pm m$ is the total number of atoms in the system after including defects. $E_{perfect}$ is the energy per atom in the perfect matrix. After MS relaxation, the MD simulations are performed at a given temperature (0 K–700 K) in which the velocity scaling method is used. The 700 K is selected not only because it is close to the operating temperatures of typical commercial fission reactors but also because it is the maximum temperature at which the effect of magnetic disorder can be neglected for BCC iron system [37]. For all calculations, the NPT ensemble is applied with zero pressure. The time-step is 1 fs and the total simulation time for each case is up to 100 ps. The elastic constants are then calculated after above full relaxations at different temperatures. The state obtained from NPT is then set as initial state for further 20,000 MD steps relaxation under NVT ensemble. The relaxations from the last 1000 MD steps are used for elastic constant calculation at different temperatures with equation (1) to ensure the statistic accuracy. Further relaxations with a longer simulation time under NVT ensemble and more steps for averaging the elastic constants have also been performed. The results show that the 20,000 MD steps of relaxation and 1000 steps of averaging is enough to get accurate results of elastic constants with a statistic uncertainty less than 1.0×10^{-4} for these three elastic constants in the following calculations.

3. Results

The elastic constants, C_{11}, C_{12}, and C_{44}, of perfect matrix at different temperatures are firstly calculated as a reference. These results are shown in Fig. 1. These elastic constants decrease with increasing temperatures, consistent with experimental results [38,39]. In other materials (e.g. Si [40] and Au [41]), a similar trend is also observed. The fitted functions for the dependence of elastic constants on temperature are:

$$C_{11} = -0.04936T + 242.0 \quad (3)$$

$$C_{12} = 3.18591 \times 10^{-5}T^2 - 0.05979T + 144.7 \quad (4)$$

$$C_{44} = 4.87617 \times 10^{-5}T^2 - 0.06903T + 116.4 \quad (5)$$

The standard errors of the fitted parameters in linear function