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Phase-field modeling of void anisotropic growth behavior in irradiated zirconium

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ABSTRACT

A three-dimensional phase-field model was developed to study the effects of surface energy and diffusivity anisotropy on void growth behavior in irradiated Zr. The gamma surface energy function used in the phase-field model was developed with the surface energy anisotropy calculated from the molecular dynamics simulations. The model assumes that vacancies have larger mobility in the *c*-axis than in the *a*-axis and *b*-axis, whereas interstitials have larger mobility in the basal plane than in the *c*-axis. The equilibrium void morphology and the effect of defect concentrations and mobility anisotropy on void growth behavior were simulated using the developed model. The simulations demonstrated that (1) the developed phase-field model correctly reproduces the faceted void morphology predicted by the Wulff construction; (2) with isotropic diffusivity, the void prefers to grow on the basal plane; and (3) when the vacancy has large mobility along the *c*-axis and interstitial has a large mobility on the basal plane of hexagonal closed-packed Zr alloys, a platelet void grows in the *c*-direction and shrinks on the basal plane. These findings are in agreement with the observation of void morphology and growth behavior in irradiated Zr, and reveal that the strong mobility anisotropy of defects results in the anisotropic growth of voids in Zr.

1. Introduction

Zr-based alloys have excellent void swelling resistance making them the principal structural materials for fuel cladding and core components in reactors [1,2]. Extensive experiments on void swelling resistance of Zr and Zr-based alloys were conducted since the 1970s. Research revealed that the void swelling behavior of Zr-based alloys depends on a variety of factors, such as radiation fluency, temperature, dislocation, precipitates, the anisotropy of materials properties, and stress states [3–10]. Anisotropy of materials properties, such as anisotropic thermal expansion [10], diffusivity [10–12], and surface or interface energy [13–16], have significant influence on the morphology of void and gas bubble and their growth kinetics in hexagonal crystals during irradiation. An anisotropic growth behavior of void, nucleating as thin platelets, growing in the *c*-direction, and shrinking along the *a*-directions with increasing dose, was observed in irradiated Zr [14]. Griffiths et al. [14,17] suggested that the anisotropic growth attributes to the diffusion anisotropy difference (DAD) between vacancies and self-interstitials. However, the mechanism of anisotropic growth is still not well understood because of the complexity of potential multi-physics coupling. Fundamental understanding of the void growth mechanism and kinetics is important in predicting the swelling kinetics in irradiated Zr-alloys, providing guidance in the design of advanced swelling resistant materials.

Computational simulation has more advantages in identifying the effects of individual and coupling physic processes on microstructure evolution kinetics compared with the experimental investigation of complicated microstructure evolution. Mesoscale phase-field approach is one of the powerful simulation tools applied to predict 3D microstructure evolution in many vital material processes, such as solidification [18–20], precipitate growth [21–23], martensitic transformations [24,25], grain growth [26,27], and growth process of bubbles and voids during irradiation [28–33]. The phase-field approach is based on thermodynamic and kinetic properties, such as defect formation energy, chemical free

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energy of coexisting phases in the system, elastic properties, anisotropic interfacial energy, and defect and interface mobility. The thermodynamic and kinetic properties can be obtained from experiments and atomistic simulations [34–36].

In this work, a phase-field model is developed to study the effects of multi-physics coupling on void growth behavior in Zr alloys. Surface energies and defects formation energies in hexagonal closed-packed (hcp) Zr single crystal were calculated using molecular dynamics (MD). Anisotropic surface energy functional and anisotropic mobility of defects are constructed with the MD results and experiments. The paper is organized as follows: in Section 2, calculation methods and models are presented, including MD calculations of surface energies and defects formation energies, the construction of anisotropic surface energy functional and anisotropic mobility of defects, and an elaborate description of the phase-field model of void evolution; in Section 3, results together with their implications are provided; in Section 4, the findings are summarized.

2. Methodology

2.1. MD calculations

Classical MD method was employed to calculate the surface and formation energies of defects (i.e., vacancies and self-interstitials) in hcp Zr single crystal. Our home developed MD package MOASP was used in the simulations. The embedded atom method (EAM) potential [37] was adopted to describe the atomic interaction in Zr. A non-orthogonal simulation cell was generated for calculating both defect formation and surface energies under periodic boundary conditions.

2.1.1. Defect formation energies

The calculations of defect formation energies were conducted under constant volume and temperature (NVT). The temperature is set as 700 K. For vacancy formation energy calculations, a simulation supercell of an hcp Zr single crystal with $(n_1a \times n_2a \times n_3c)$ was generated. a and c are lattice constants set as 3.2312 Å and 5.1477 Å, respectively. The simulation supercell is initially relaxed under constant pressure and temperature (NPT) [38]. Then, one Zr atom near the box center is removed to form a vacancy. The vacancy formation energy E_v^f is calculated as:

$$E_v^f = E - (N-1) \times \frac{E_0}{N} \quad (1)$$

where E_0 is the total energy of the perfect system with N Zr atoms; and E is the total energy of the system including $(N-1)$ Zr atoms and one vacancy.

The stability of interstitials is considered in calculating self-interstitial formation energies because eight potential interstitial sites exist in the lattice of Zr crystal. According to Ref. [37], the Octahedral (O) configuration is the most stabilized configuration on interstitial defects. The self-interstitial formation energy was obtained from the O configuration in the current work. The calculation of interstitial formation energy E_i^f is similar to vacancy formation energy which is calculated as:

$$E_i^f = E' - (N+1) \times \frac{E_0}{N} \quad (2)$$

where E' is the total energy of the system including one self-interstitial.

In the simulation, a supercell $(20a \times 20a \times 20c)$ with 16,000 ± 1 Zr atoms was used. The results are summarized in Table 1. A variation of the defect formation energies calculated with different fidelity and interatomic potentials are observed. However, the tendency that the formation energy of an interstitial is larger than that of a vacancy in hcp Zr is the same. This work aims to develop the thermodynamic models for more quantitative phase-field simulations by using the thermodynamic properties from atomistic simulations.

2.1.2. Surface energies

NVT ensemble with a Langevin thermostat [38] was used for the calculation of surface energies. Five simulation cells with different hcp Zr-vacuum interfaces were constructed to characterize the surface energy anisotropy of hcp Zr. The initial velocity of Zr atoms in the simulation cells was generated according to the Boltzmann distribution for a given temperature. The entire simulation cell was allowed to relax in the normal direction of the surface for 200 ps. The non-orthogonal simulation cell for the calculation of $(10\bar{1}0)$ surface energy is shown in Fig. 1. A supercell with N Zr atoms was built, and then a layer of vacuum with a

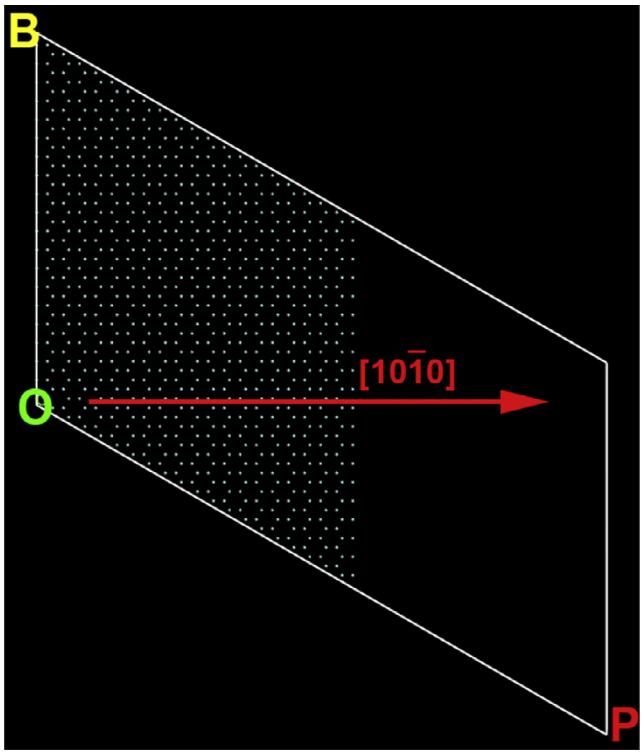

Fig. 1. An illustration of the simulation cell and the normal direction of the surface $(10\bar{1}0)$ for the calculation of $(10\bar{1}0)$ surface energy.

Table 1
MD calculations of defect formation energies in Zr.

Defect formation energies	This work (eV)	Ref.		
		[39] (eV)	[40] (eV)	[41] (eV)
E_v^f	2.024	1.59	1.79	1.74
E_i^f	2.971	3.92	4.005	2.75