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Nuclear Instruments and Methods in Physics Research B 255 (2007) 52–56
NIM B
Beam Interactions with Materials & Atoms
www.elsevier.com/locate/nimb

Modeling of He-bubble migration in bcc Fe

Kazunori Morishita *, Ryuichiro Sugano

Institute of Advanced Energy, Kyoto University, Gokasho, Uji, Kyoto 611-0011, Japan

Available online 11 December 2006

Abstract

Kinetic Monte-Carlo simulations were performed to investigate the migration of He-bubbles in bcc Fe, using the binding energies of point defects to a He-bubble and the migration energies of an Fe atom on bubble surface, both of which are obtained by molecular dynamics and molecular static calculations. This approach linking multiple simulation methods can remove the degrees of freedom for lattice vibrations effectively and enables us to perform long time scale simulations successfully. When the equilibrium concentration of point defects in a matrix is assumed, a He-bubble can migrate by the surface diffusion mechanism and show the Brownian motion. The diffusion coefficient of larger He-bubbles than about 0.8 nm in diameter is proportional to d^{-4}, where d is bubble diameter. It is in good agreement with conventional continuum theory predictions.

PACS: 61.80.Az; 61.72.Ji; 67.80.Mg; 66.30.−h

Keywords: He-bubble; Bubble migration; Fe; KMC simulation; Multiscale modeling

1. Introduction

Radiation damage processes [1] are in essence multiscale phenomena, which occur over a wide spectrum of time and length scales. It is very difficult to evaluate the processes by experimental techniques in particular at relatively small time and length scales. Hence, many challenges have been made to understand these processes by computer simulation techniques. However, even if computational techniques are available, the simulation time and length scales are limited. For example, a molecular dynamics (MD) technique is a powerful tool to investigate displacement cascades in materials, but its simulation time is limited to 100 ps at the most. Therefore, linking various simulation techniques is required to overcome the scale limit that individual techniques inevitably have.

He-bubbles are three-dimensional vacancy clusters containing He atoms, and are known to migrate in materials [2–6]; however, bubble diffusivity is so low that the time scale of the diffusion is, in many cases, beyond the limit of the MD method. This paper describes a valuable attempt to evaluate the diffusivity of He-bubbles in bcc Fe by a kinetic Monte-Carlo (KMC) simulation technique that employs defect energies obtained using MD and molecular static (MS) calculations.

2. KMC model

It is known that He-bubble movements in solids occur by the following two mechanisms. One is the so-called *volume diffusion mechanism*, where a He-bubble moves each time it absorbs or emits point defects such as vacancies and self-interstitial atoms (SIAs). The influx of point defects to a spherical He-bubble is given by $\nu^k_{\text{influx}} = 4\pi R D_k C_k/\Omega$ in the unit of s^{-1}, where D_k and C_k are the diffusion coefficient and concentration of the type k point defect in the matrix, respectively, where k denotes vacancies, self-interstitial atoms (SIAs) and interstitial He atoms. Ω is atomic volume and R is bubble radius. Note that, bubble volume is given by $V = N_V\Omega = 4\pi R^3/3$, where N_V is the number of vacancies contained in the bubble.

* Corresponding author. Tel.: +81 774 38 3477; fax: +81 774 38 3479.
E-mail address: morishita@iae.kyoto-u.ac.jp (K. Morishita).

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doi:10.1016/j.nimb.2006.11.010

This equation may show a connection between He-bubbles characterized by radius R from the continuum model viewpoint and those characterized by N_V from the atomistic model viewpoint. The migration energy of point defects in an unstrained matrix is 0.74, 0.058 and 0.078 eV for a vacancy, an SIA and an interstitial He atom, respectively [7]. The concentration of point defects in the matrix during *irradiation* is usually determined by solving simultaneous rate theory equations, which describe various defect interactions such as production, recombination, absorption by sinks and emission from sinks. In the present study, however, it was just given by the equilibrium condition, i.e. $C_k = \exp(-E_k^F/k_B T)$, where E_k^F is the formation energy of the type k point defect, k_B is the Boltzmann constant and T is temperature. The formation energy of point defects employed here is 1.70, 4.88 and 5.25 eV for a vacancy, an SIA and interstitial He, respectively [7]. The outflux of point defects from a He-bubble was written by $v_{\text{outflux}}^k = (4\pi R D_k/\Omega)\exp(-E_k^{\text{bind}}/k_B T)$. Here, E_k^{bind} is the binding energy of point defects to the He-bubble, which was already obtained by MD and MS techniques [7–10] and by a continuum model approach [11], as a function of the He-to-vacancy (He/V) ratio of bubbles.

The other is the so-called *surface diffusion mechanism*, where He-bubble movements in solids are accompanied with the diffusion of matrix metal atoms on bubble surface. The earlier continuum model describing this mechanism [2,12] indicates that an analytical expression of the diffusion coefficient of He-bubbles is inversely proportional to their radius to the fourth power. In the present study, an activation energy for Fe atom jumps on bubble surface was assumed to be described by $Q = \Delta E + E_m$ as depicted in Fig. 1 and atom jump frequency was given by $\nu_{\text{surf}} = \nu_0 \exp(-Q/k_B T)$, where ν_0 was assumed to be 10^{13} s. ΔE denotes a difference in total potential energies between an Fe atom at stable sites before and after a possible jump and was given by:

$$\Delta E = \begin{cases} E_{\text{pot}}^{\text{after}} - E_{\text{pot}}^{\text{before}} & \text{if} \quad E_{\text{pot}}^{\text{after}} - E_{\text{pot}}^{\text{before}} > 0 \\ 0 & \text{if} \quad E_{\text{pot}}^{\text{after}} - E_{\text{pot}}^{\text{before}} \leqslant 0 \end{cases}. \tag{1}$$

The potential energy E_{pot} was assumed to be a function of the Fe coordination number of Fe atoms and the He/V ratio of bubbles, where the Fe coordination number of Fe atoms was defined by the number of Fe atoms within the first nearest neighbor distance from a particular Fe atom. For example, the Fe coordination number of an Fe atom neighboring a vacancy in the bcc crystal is 7. The relationship between potential energies and the Fe coordination number was roughly obtained using MS calculations with various Fe *flat* surfaces where interatomic potential sets employed were the same as our previous work [7–10]. Calculations were done with and without He atoms. In the former case, He atoms were introduced into a vacant space above the flat surface so that the He/V ratio of the space equals to 1; and in the latter case there was a perfect vacuum above the surface. The results are shown in Fig. 2. The potential energy does not depend much on He/V ratios in the investigated range, which is empirically described as a linear function of the Fe coordination number and He/V ratios, as follows:

$$E_{\text{pot}} = -2.3568 - 0.2449M + (0.3389 - 0.02999M) \times (\text{He/V}), \tag{2}$$

where M denotes the Fe coordination number and E_{pot} is given in the unit of eV. This equation will be used when He/V ratios are less than or equal to 1. Fig. 3 shows the formation energy of an empty void calculated by the Fe coordination number of Fe atoms on void surface with Eq. (2). As a reference, a relaxed formation energy obtained by MD and MS calculations is also plotted in the figure, which is well fitted to $E_{\text{void}}^F = 2.79N_V^{2/3} - 0.755N_V^{1/3}$. The figure indicates that the formation energy evaluated using Eq. (2) is very consistent with the MD and MS results, although there seems to have some differences at relatively large size. This small difference is probably caused by a difference between the two evaluation methods. Namely, the present method only considers the contribution of the

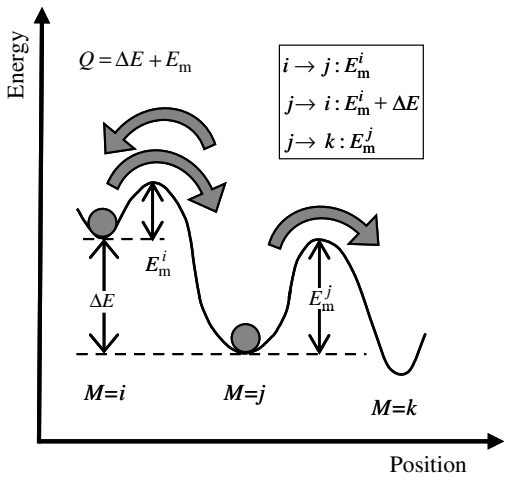

Fig. 1. Schematic representation of activation energies for Fe atom jumps on surface.

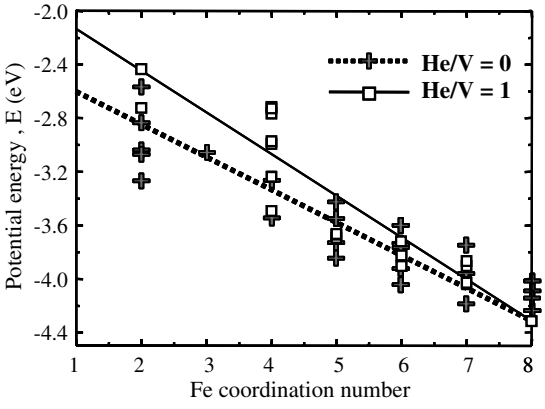

Fig. 2. Atom potential energies as a function of Fe coordination numbers and He/V ratios.