Computational Materials Science 156 (2019) 268–272

Contents lists available at ScienceDirect

Computational Materials Science

journal homepage: www.elsevier.com/locate/commatsci

Development of a pair potential for Ta-He system

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ARTICLE INFO

Keywords:
Tantalum
Helium
Pair potential

ABSTRACT

A pair potential for Ta-He system was developed by fitting to the results obtained from *ab initio* calculations. The potential model proposed by Juslin and Nordlund was employed to describe the Ta-He interaction. The formation energies of single He atom at different sites were utilized as the fitting targets. Particle swarm optimization scheme was adopted to determine the parameters. The newly developed potential could reproduce the formation energies of single He defects very well. Besides, the binding energies of an additional interstitial He atom to an existing $He_{n-1}V$ and He_n clusters, and the migration energies of interstitial He atom and HeV_2 cluster were studied. They were found to be in good agreement with available *ab initio* results.

1. Introduction

Helium (He), which is generated by (n, α) transmutation reactions under 14 MeV neutron irradiation, plays a significant role in microstructural evolution and mechanical properties degradation [1,2]. Since the solubility of He atoms in metals is very low, it is effortless for He atoms to aggregate and generate helium bubbles, which accelerates irradiation-induced swelling and embrittlement, particularly at high temperature [3]. Besides, due to the high mobility of He atoms via the interstitial migration mechanism and the strong binding with vacancies [4], it is of great difficulty to explore the atomistic behavior experimentally. Many researchers have tried studying these properties by performing *ab initio* calculations. Simple He defects in metals have been investigated systematically [5,6]. However, the high computational cost of *ab initio* calculations makes it limited in researching systems with only hundreds of atoms, which is generally not sufficient for investigating the nucleation of helium bubbles or similar behaviors. From this perspective, molecular dynamics (MD) and Monte Carlo (MC) simulations are the most practical approaches currently available to study He behaviors in metals on desired scale.

For both MD and MC simulations, the interatomic potential, which describes the interactions among atoms, is one of the most predominant factors determining both the efficiency of the computational process and the accuracy of the results [7]. To explore the interactions between He and individual metals, various interatomic potentials have been developed in the past years and many works in depth have been done based on these potentials [8–16]. As a typical body centered cubic (BCC) transition metal, tantalum (Ta) plays an important role in nuclear materials [17–19]. Thus Ta and its alloys have been investigated using both the experimental and *an initio* methods [5,6,20–25]. The modeling of He defects in Ta matrix using MD simulation can be traced back to decades ago. In early, Wilson et al. [26] proposed a pair potential for Ta-He system based on the Wedepohl method [27] and researched some basic He defect energies in Ta matrix. However, in these calculations, pair potential for Ta-Ta interaction was used, which might be inadequate to capture the lattice response of the metals after He defects are introduced. More importantly, the He-He interaction was not included, therefore the modeling of He clusters or bubbles was not feasible. Besides, *ab initio* calculations have demonstrated that He atoms tend to occupy the tetrahedral interstitial sites compared with octahedral ones, while Wilson's potential can't predict this clearly [6,28].

In present work, a new potential for Ta-He system based on the model proposed by Juslin and Nordlund [9] is presented. Combining with the embedded atom method (EAM) potential [29] for Ta-Ta interactions and the Hartree-Fock-Dispersion pair potential [30] for He-He interactions, the formation energies of single He defects can be

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https://doi.org/10.1016/j.commatsci.2018.09.057
Received 5 February 2018; Received in revised form 28 August 2018; Accepted 30 September 2018

reproduced in good agreement with *ab initio* results. Furthermore, the binding energies of an additional interstitial He atom to $He_{n-1}V$ and He_n clusters and the migration energies of interstitial He atom and HeV_2 clusters have been investigated and found to be in good agreement with available *ab initio* results.

2. Methodology

2.1. Definitions

For the convenience of analysis and discussion later, several definitions used in the following context were given out firstly. He_nV cluster is defined as n He atoms located in one Ta vacancy and He_n cluster is defined as n He atoms located in one Ta interstitial site. The formation energy of He_nV cluster or He_n cluster is defined as:

$$E_f(He_nV/He_n) = E_{total} - N_{Ta}E_{Ta} - N_{He}E_{He}, \quad (1)$$

where E_{total} is the total energy of a system containing the He_nV or He_n cluster, N_{Ta} and N_{He} are the numbers of Ta atoms and He atoms in the system, E_{Ta} is the cohesive energy of BCC Ta and E_{He} is the energy of an isolated He atom, respectively. For MD simulations, E_{He} is regarded as 0. Since the He atoms could be located in both the tetrahedral interstitial sites and the octahedral interstitial sites, there are two different formation energies of interstitial He atoms, expressed as E_f^{Tetra} and E_f^{Octa}, respectively. The binding energy of an additional interstitial He atom to the He_nV cluster, $E_b(He, He_nV)$, is defined as:

$$E_b(He, He_nV) = E_f(He) + E_f(He_{n-1}V) - E_f(He_nV), \quad (2)$$

where $E_f(He)$ is the formation energy of a single tetrahedral interstitial He in perfect Ta crystal. Similarly, the binding energy of an additional interstitial He atom to an existing He interstitial cluster He_n, $E_b(He, He_n)$, could be calculated by:

$$E_b(He, He_n) = E_f(He) + E_f(He_n) - E_f(He_{n+1}). \quad (3)$$

2.2. Computational methods

Since it is difficult to obtain the energetics of He defects and small clusters in BCC Ta matrix from experiments, which are important input parameters for fitting interatomic potential, the results from *ab initio* calculations in the framework of density functional theory (DFT) were employed. To the best of our knowledge, there are some available values based on DFT calculations reported in literatures [5,6]. However, they are not sufficient for present work. To guarantee the consistency of the calculated values, we calculated all the relevant values by performing DFT calculations, which were used for fitting the potential parameters of Ta-He. Besides, to verify the accuracy of the newly developed potential, we also calculated the available values by DFT calculations correspondingly as a comparison.

All the DFT calculations in present work were performed using the Vienna *ab initio* simulation package (VASP) [31]. Exchange and correlation were treated at the Perdew-Burke-Erzernhof (PBE) functional level [32]. The calculations were performed using supercell models of the defect structures in which He atoms were placed at the appropriate positions of a large cell that was a $4 \times 4 \times 4$ repeat of the underlying BCC structure with the equilibrium lattice parameter. Then, the atomic coordinates and volume were relaxed to minimize the forces and pressure. Brillouin-zone sampling was performed using the Monkhorst-Pack scheme [33] with a $3 \times 3 \times 3$ k-point mesh. The energy tolerance for self-consistent field convergence was 10^{-5} eV/atom with the plane ware energy cutoff of 500 eV. The equilibrium lattice parameter obtained for Ta based on DFT calculations in present work is 3.3089 Å, which agrees well the experimental data 3.3013 Å [34]. The formation energies of vacancy, substitutional He and interstitial He are listed in Table 1, in comparison with related literatures and those calculated by using generalized gradient approximation (GGA) of Perdew and Wang (PW91) [35]. As to the formation energy of substitutional He, the values subtracted by corresponding vacancy formation energy were listed in the parentheses. From the table, it can be seen that due to the utilization of different pseudo-potentials and parameters, there exist slight differences among the DFT results. Despite that, most values agree well (except the vacancy formation energy from Ref. [6], which is much larger than the others.).

Table 1
Formation energies of vacancy, substitutional He and interstitial He in BCC Ta matrix. All values are in eV. The values in the parentheses are the formation energies of substitutional He subtracted by corresponding vacancy formation energies.

		Vac.	Sub.	Octa.	Tetra.
DFT	Ref. [5]-PW91	2.86	4.61 (1.75)	3.42	3.16
	Ref. [6]-PW91	3.27	4.58 (1.31)	3.65	3.34
	Present-PW91	2.80	4.56 (1.76)	3.69	3.36
	Present-PBE	2.88	4.68 (1.80)	3.74	3.41
MD	Wilson	–	– (0.93)	4.23	4.22
	Present	2.98	4.68 (1.83)	3.57	3.41

All the MD simulations in present work were performed using the software package LAMMPS [36]. The simulations were performed in a $10 \times 10 \times 10$ supercell of conventional BCC supercell with the equilibrium lattice parameter.

2.3. Parameterization of potentials

As mentioned in Section 1 briefly, in our MD simulations, the embedded atom method (EAM) potential proposed by Zhou et al. [29] was used to describe the interactions of Ta-Ta. The pair potential proposed by Aziz [30] was taken for He-He interactions. To check the accuracy of the potentials, the cohesive energies of perfect BCC Ta crystal, E_c(Ta), and of perfect FCC He crystal, E_c(He) were calculated to be 8.09 eV/atom and −0.00714 eV/atom, respectively, equalling to the corresponding experimental values [37,38], which validates the accuracy of the potentials for Ta-Ta and He-He.

Although many-body potential models have been used in describing the interactions between He and metals [8], Juslin and Nordlund pointed out that pair potential should be enough since He is a noble gas element [9]. Besides, pair potentials are with superiority in efficiency compared with many-body potentials, which makes it available for exploring larger systems. Hence, pair potential was taken to describe the interaction of Ta-He in present work. Specially, the pair potential model (JN-model) proposed by Juslin and Nordlund [9], which has been applied to describe the interactions of Fe-He [9], Cr-He [13] and W-He [16] successfully, was adopted in present work. The formulation of JN-model is given as:

$$f(r_{ij}) = \begin{cases} \text{ZBL}, & r_{ij} \leqslant r_1, \\ p_3 r_{ij}^3 + p_2 r_{ij}^2 + p_1 r_{ij}^1 + p_0, & r_1 \leqslant r_{ij} \leqslant r_2, \\ \left(a + \frac{b}{r_{ij}}\right) e^{-cr_{ij}} f_c(r_{ij}), & r_{ij} \geqslant r_2, \end{cases} \quad (4)$$

where ZBL represents Ziegler-Biersack-Littmark function [39], which is often used to describe the short-range interaction, and f_c is the cut-off function, given by

$$f_c(r_{ij}) = \begin{cases} 1, & r_{ij} \leqslant r_c - r_d, \\ \frac{1}{2}\left(1 - \sin\frac{\pi(r_{ij} - r_c)}{2r_d}\right), & |r_c - r_{ij}| \leqslant r_d, \\ 0, & r_{ij} \geqslant r_c + r_d. \end{cases} \quad (5)$$

In Eqs. (4) and (5), a, b, c, p_h (h= 0 ~ 3), r_1, r_2, r_c, r_d are parameters to be determined in JN-model. In JN-model, the first section comes from ZBL function or DMOL [40] calculations directly. The third section is the most crucial for describing the interaction between He and