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Development of Fe-C interatomic potential for carbon impurities in α-iron

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ABSTRACT

Two versions of interatomic potentials for iron-carbon system based on the analytic bond-order potential formalism were developed and tested on diffusion of carbon in body-centered cubic (BCC) iron. By using these potentials, we found that carbon diffuses from octahedral site to the next nearest neighbor octahedral site via tetrahedral site, with energy barriers of 0.74 and 0.70 eV, respectively. These values are in good agreement with the transition from octahedral site to octahedral site via tetrahedral site with energy barriers of about 0.81–0.87 eV, observed in experiment. Other theoretical works found similar diffusion mechanism with energy barriers of about 0.72–0.92 eV. We also verified the transferability of the newly developed potential in the study of the effect of carbon on the body-centered cubic/face-centered cubic (BCC/FCC) phase transition in iron following the phase transition in the Bain mechanism. We found that the presence of carbon leads to the decrease of the BCC to FCC transformation energy barrier, consistent with first-principles calculations. Thus, the new potentials can be good candidates for modeling carbon diffusion and the transformation processes in iron-carbon alloys.

1. Introduction

Materials design, which could simultaneously demonstrate multiple promising properties like high strength, good ductility and fracture toughness, has been a long-standing demand for industrial applications. Carbon steel, which usually consists of ferrite and cementite phases at room temperature, is one of the most popular engineering structural materials. It is well known that carbon is a very important impurity in the iron matrix and can affect significantly the mechanical properties of iron/steel structures. The distribution and diffusion of carbon in iron can control the formation and kinetics of many processes in steels such as carbide precipitation, martensite ageing, and ferrite transformation [1–3], which are of major importance for the steel industry. Traditionally, for normal steel, as the carbon content increases, the hardness of the steel also increases while the ductility decreases and vice versa. This means that there is a trade-off between the strength and ductility. Since the mechanical properties of materials depend on microstructure [4], to obtain both strength and ductility properties, one considers to control the microstructure via grain refinement, precipitation control, and so on. Recently, due to the improvement of experimental techniques, many innovative methods have been realized. One is to use heterogeneous steel such as dual-phase (DP) steel, which consists of ferrite and a dispersed hard martensitic second phase in the form of islands [5]. Depending on the carbon content and the morphology of the microstructure, DP steel can exhibit a number of advantageous properties compared to other steels, for example, excellent resistance to fatigue crack propagation at growth rates close to fatigue threshold. In spite of that, the understanding about the transformation dynamics of DP steel, the effect of carbon solute and the formation of carbide in DP steel are still lacking.

In order to study deeply the role of carbon and the microscopic phenomena involving carbon in steel, atomistic modelling at large time and length scales are essential, aside from experimental measurements. While first-principles methods such as Density Functional Theory (DFT) are rapidly growing and proved to give considerable accuracy, the simulation models are limited to few hundreds of atoms because these methods explicitly describe the electron-electron/electron-nucleus interactions at quantum level [6]. To fulfill the simulations at large time/length scales, alternative methods in conjunction with empirical potentials such as Molecular Dynamics or Kinetic Monte Carlo can be good choices. In these computational methods, accurate interatomic potentials are usually required to find the total energy and the equilibrium state of the system under investigation. There have been several works on the development of iron-carbon (Fe-C) interatomic potentials for

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modelling Fe-C alloys. However, due to the complexity of Fe-C bonding structure, these potentials have their limitations [7–9]. For instance, Ruda et al. [9] developed an interatomic potential for Fe-C system based on the Embedded Atom Method (EAM) [10] which reproduced the equilibrium lattice constants, bulk moduli, and cohesive energies for several carbide structures but it finds the tetrahedral site (T-site) as the most stable site. This is opposite to the first-principles results where the octahedral site (O-site) is found to be more stable. The reason may come from the choice of the potential formalism because the EAM potential has central-force characteristic, which is not good for describing non-metallic elements. To include different bonding types as in case of Fe-C alloy, a more extensive potential framework should be used. One of them is the Modified Embedded Atom Method (MEAM) potential created by Baskes [11], in which the EAM was modified to include the directionality of bonding. Some potentials based on this formalism have been developed for investigating the interactions between carbon interstitial solute atoms with various defects in iron as vacancies, dislocations and grain boundaries [12], as well as the properties of cementite (Fe_3C) [13]. Another framework is the analytic bond-order potential (ABOP) formalism [14], which is constructed based on the modification of the Brenner and Tersoff potentials [15–19] and found to be suitable for metallic/non-metallic compounds [20–24]. As compared to the MEAM potential, the ABOP was reported to consume much less computational time and memory [25]. For a small system, this difference is negligible. However, for a large simulated system, it can be a significant impact on the simulation speed. Furthermore, it has been found that the ABOP potential for pure iron simultaneously shows excellent descriptions of iron both in the BCC and FCC phases including the phonon dispersion curves. It also properly reproduces the α–γ–δ transition sequence [26]. Therefore, the use of the ABOP formalism for constructing Fe-C potential will be conducted in the present work. So far, one interatomic potential for Fe-C system based on the ABOP formalism has been developed and proved to give a good description of the basic physical properties of several cementite structures such as Fe_3C, Hägg carbide Fe_5C_2, and Eckstrom–Adcock carbide Fe_7C_3 [24]. However, it fails to reproduce the correct diffusion path of single carbon from O-site to the first nearest neighbor O-site. As we can see in Fig. 1, this potential produces a quite high energy barrier for carbon diffusion and, more importantly, a local minimum in the minimum energy path (MEP) of the carbon diffusion process, which is in contradiction with the result from Density Functional Theory calculation. Thus, it is necessary to construct a new interatomic potential for Fe-C system, which can correctly describe the dynamical process of carbon as well as the effect of carbon on the BCC/FCC transformation in iron/steel.

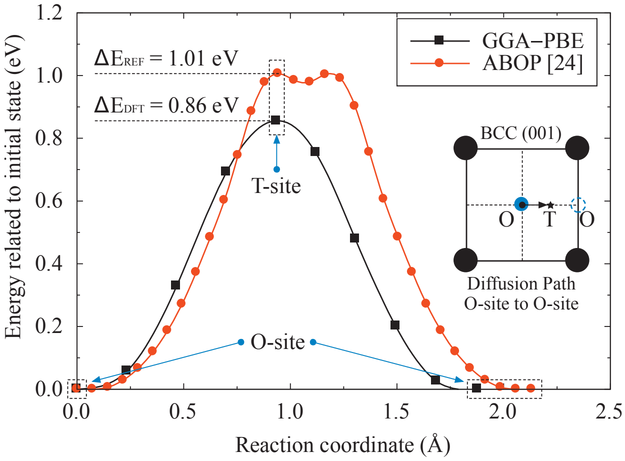

Fig. 1. Diffusion paths and energy barriers of carbon from O-site to the first nearest neighbor O-site by first-principles (black curve) and analytic bond-order potential [24] (red curve) calculations using Climbing Image Nudge Elastic Band method. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

In this work, the interatomic potential for Fe-C system was developed by fitting the potential parameters to the energies and atomic forces from first-principles calculations by using Genetic Algorithm as optimization method. The details of the paper will be as follows: First, the ABOP formalism will be introduced. Secondly, the database for fitting and the optimization processes will be discussed. Finally, the potential parameters will be presented and validated with first-principles calculations.

2. Methodology

2.1. Potential formalism

The potential formalism originates from the concept of bond order potential proposed by Pauling [27], which states that the strength of a bond between two atoms is not constant, but depends on the local environment, i.e. an atom with many neighbors forms weaker bonds than an atom with few neighbors. It was calibrated firstly for silicon and later for carbon [18,19]. In the form of Albe et al. [20,28], the total potential energy is written as:

$$E = \frac{1}{2}\sum_{i\neq j} V_{ij} = \frac{1}{2}\sum_{i\neq j} f_{ij}^{C}(r_{ij})[V_{ij}^{R}(r_{ij}) - b_{ij}V_{ij}^{A}(r_{ij})] \tag{1}$$

where $V^R(r)$, $V^A(r)$ are repulsive and attractive Morse-type pair potentials, respectively:

$$V^R(r) = \frac{D_0}{S-1}\exp(-\beta\sqrt{2S}(r-r_0)) \tag{2}$$

$$V^A(r) = \frac{SD_0}{S-1}\exp(-\beta\sqrt{2/S}(r-r_0)) \tag{3}$$

and $f^C(r)$ is the cut-off function to restrict the interaction range:

$$f^C(r) = \begin{cases} 1, & r \leqslant R-D \\ \frac{1}{2} - \frac{1}{2}\sin\left(\frac{\pi}{2}\frac{r-R}{D}\right), & R-D < r \leqslant R+D \\ 0, & r > R+D \end{cases} \tag{4}$$

In Eqs. (2) and (3), D_0 is the dimer bond energy, r_0 is the dimer bond distance, S is an adjustable parameter while β can be determined from the ground-state oscillation frequency of the dimer:

$$\beta = k\frac{2\pi c}{\sqrt{2D_0/\mu}} \tag{5}$$

where k is the wave vector and μ is the reduced mass of the dimer.

For the cut-off function, R and D are adjustable parameters in which $R + D$ is the cut-off distance while $R-D$ is the distance where the potential starts to vanish.

The important feature of this potential is the presence of the b_{ij} term, a bond-order term, which includes three-body interactions and angularity:

$$b_{ij} = (1 + \chi_{ij})^{-1/2} \tag{6}$$

where:

$$\chi_{ij} = \sum_{k\neq i,j} f_{ik}^{C}(r_{ik})g_{ik}(\theta_{jik})\omega_{ijk}\exp(\alpha_{ijk}(r_{ij}-r_{ik})) \tag{7}$$

defines the effective coordination number of atom i, i.e. the number of nearest neighbors, taken into account the relative distance of two neighbors r_{ij}–r_{ik} and the bond-angle θ_{jik}. Here, ω_{ijk} and α_{ijk} are fitting parameters depending on the types of the atoms in the triplet i–j–k. These are added to make the bond order term, b_{ij}, compatible to the expression of Brenner for hydrocarbon [15,16]. In the case of Fe-C, these parameters are set to 1 and 0 for ω_{ijk} and α_{ijk}, respectively.

In Eq. (7), the angular function $g(\theta)$, depends on the angle θ