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An improved modified embedded-atom method potential to fit the properties of silicon at high temperature

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ABSTRACT

An improved modified embedded-atom method (MEAM) potential for Si was optimized. The potential could quite well fit the potential energy curve and the elastic constants of diamond Si, the pair correlation function, the structure factor and the bond angle distribution function of liquid Si. The potential could also well fit the melting point and the solid-liquid phase transformation enthalpy. The potential could be used to study the solid-liquid transformation behaviors and the liquid structure of Si, and it could also be used to develop a MEAM potential for the binary Al-Si alloy.

1. Introduction

Si is an important semiconductor material and the main adding element of many alloys, and its macroscopic property is closely related to its structural and dynamic characteristics on the atomic scale. The molecular dynamics (MD) simulation based on the empirical potential is an important way to study the behavior of Si on the atomic scale. However, the accuracy of the MD simulation is dependent on the quality of the interatomic potential.

By including the angle-dependent electron density, the modified embedded-atom method (MEAM) potential is able to describe the metal, the semi-metal and the non-metal systems [1–3]. Therefore it is ideal to use MEAM model to describe the Si and the Si-containing system. More than ten MEAM potentials for Si have been developed [4–13]. However, the transition from covalent bonding to metallic bonding occurs during the solid-liquid phase transformation of Si [14], which results in a large deviation of the melting point, the solid-liquid phase transformation enthalpy and the liquid structure properties calculated by these MEAM potentials. For example, Liu et al. [13] compared the melting points of Si calculated by different MEAM potentials, and the melting points given by Lee [8], Cui [11], Jelinek [12] and Liu [13] potential were 2930 K, 2325 K, 1385 K and 1295 K, which had a large deviation from the experimental value of 1685 K [9]. Among the all MEAM potentials for Si, only the Ryu potential [9] could well fit the solid-liquid transformation properties of Si. However, as the pair correlation function calculated by the Ryu potential [9] was far away from the density function theory (DFT) curve [15], the Ryu potential could not be used to describe the liquid structure of Si. In total, the current MEAM potentials could not well describe the properties of Si at high temperature.

The liquid structure of Al-Si alloy is a long-standing issue [16–20] and MD simulation is a promising method to further study it. As the current MEAM potential for Si could not well describe the high temperature properties of Si, the existing MEAM potentials [12,21] for Al-Si alloy could not be used to study the liquid structure. To develop a MEAM potential for the binary Al-Si alloy to study the liquid structure of Al-Si alloy, a MEAM potential for Si that well describe the high temperature properties of Si is also needed.

In total, the present paper is to develop a MEAM potential that could well describe the properties of Si at high temperature, including the solid-liquid transformation properties and the liquid structure.

2. Method

2.1. MEAM potential

The total energy E of a system of atoms in the MEAM is approximated as the sum of the atomic energies

$$E = \sum_i F_i(\bar{\rho}_i) + \frac{1}{2}\sum_{j\neq i} S_{ij}\phi_{ij}(r_{ij}) \quad (1)$$

where F_i is the embedding energy function, $\bar{\rho}_i$ is the background

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electron density at the site of atom i, ϕ_{ij} is the pair potential between atoms i and j separated by a distance r_{ij}, and S_{ij} is the screening factor. The pair potential could be obtained by the subtraction of the embedding energy from the total energy of the reference structure. The total energy of the reference structure could be described by the following universal equation of state (EOS) proposed by Rose et al. [2]:

$$E^u(r) = -E_c(1 + a^* + d\cdot(a^*)^3)e^{-a^*} \quad (2)$$

$$a^* = \alpha(r/r_e - 1) \quad (3)$$

$$\alpha = (9B\Omega/E_c)^{1/2} \quad (4)$$

$$d = \begin{cases} d_{att}, & a^* > 0 \\ d_{rep}, & a^* < 0 \end{cases} \quad (5)$$

where r is the nearest neighbor distance, r_e is the equilibrium nearest neighbor distance, B is the bulk modulus of the reference structure, Ω is the atomic volume, and d_{att} and d_{rep} are the adjustable parameters.

The details of the MEAM formalism were well described in literatures [1–3]. The potential parameters that determine the MEAM potential for an element are the parameter A describing the ratio between the embedding energy and the total energy, the parameters $\beta^{(0)}$, $\beta^{(1)}$, $\beta^{(2)}$, $\beta^{(3)}$, $t^{(1)}$, $t^{(2)}$ and $t^{(3)}$ describing the background electron density, the parameters E_c, a, α, d_{att} and d_{rep} describing the EOS, the parameter r_c describing the cutoff effect while calculating the electron density and pair potential, and the parameters C_{min} and C_{max} describing the many-body screening factor.

2.2. Fitting process

The fitting of the MEAM potential parameters is a process with multiple parameters and objects. The particle swarm optimization algorithm was used to optimize the potential parameters. Each particle is set an initial position (x) and an initial velocity (v), and the position is the set of the potential parameters, while the velocity is the change value of the set of the potential parameters. The position of the particle converged to the global optimal solution by continuous optimizing the position of the particles.

Before the fitting process, the constant parameters were first determined. The diamond Si was chosen as the reference structure. The cutoff r_c was set to 5 Å, which is larger than the third-nearest neighbor distance of diamond Si. By using this value, the cutoff effect is totally eliminated by many-body screening effect while calculating the electron density and pair potential. The parameters E_c, a and α were determined by the experimental value. The cohesive energies and the lattice constants of different structures of Si at 0 K, and the elastic constants of diamond Si at 0 K were used to determine the initial value and the range of the other potential parameters, which reduced the total fitting time. Further, the potential parameters were fitted by the cohesive energies of different structures of Si at 0 K, the elastic constants of diamond Si at 0 K, the melting point, the enthalpy and the volume difference of solid-liquid phase transformation, and the pair correlation function and the structure factor of liquid Si. The solid-liquid coexistence method was applied to calculate the melting point of Si. In order to reduce the fitting time, small computing model was used at the beginning of fitting, while large computing model was applied at the later stage of fitting to increase the fitting accuracy.

3. Potential parameters

The present fitted MEAM potential parameters for Si are listed in Table 1. Three representative MEAM potential [8,9,12] parameters for Si are also listed in Table 1 for comparison. The potential reported by Lee et al. [8] well fitted the enthalpy and the volume difference of the solid-liquid phase transformation, and the pair correlation function, but the calculated melting point by the potential was as high as 2930 K. The potential reported by Ryu et al. [9] improved the melting point and the solid-liquid phase transformation enthalpy, but it could not well describe the liquid structure. The potential reported by Jelinek et al. [12] was a widely-used multicomponent potential containing Si, but the properties at high temperature were not considered during fitting the potential. The present values of the parameters lat, E_c, a and α are the same as that of the reported MEAM potentials [8,9,12], as shown in Table 1. The parameters A, C_{min}, C_{max}, d_{rep} and d_{att} have effect on the properties of Si at high temperature, which were not chosen as the default values of 1, 2.0, 2.8, 0 and 0 to improve the accuracy of the potential.

Table 1
The MEAM potential parameters for Si. The unit of E_c and a is eV, and the other parameters are dimensionless.

Parameters	lat $\beta^{(3)}$	E_c $t^{(1)}$	a $t^{(2)}$	A $t^{(3)}$	α C_{min}	$\beta^{(0)}$ C_{max}	$\beta^{(1)}$ d_{rep}	$\beta^{(2)}$ d_{att}
Lee [8]	'dia' 7.5	4.63 1.8	5.43 5.25	0.58 −2.61	4.87 1.41	3.55 2.8	2.5 0	0 0
Ryu [9]	'dia' 5.5	4.63 3.13	5.43 4.47	1 −1.8	4.87 1.85	4.4 2.8	5.5 –	5.5 –
Jelinek [12]	'dia' 5.5	4.63 2.05	5.43 4.47	1 −1.8	4.87 2.0	4.4 2.8	5.5 0	5.5 0
Present	'dia' 6.05	4.63 2.09	5.43 5.16	0.90 −1.18	4.87 1.18	4.23 3.19	2.19 0.09	4.48 0.04

4. Verification of physical properties

The MEAM potential developed in this text is mainly for the description of the properties of Si at high temperature, including the solid-liquid phase transformation behavior and the liquid structure. Therefore the solid-liquid phase transformation properties and the liquid structure of Si should be carefully checked. The diffusivity of liquid Si was also checked. In addition, the solid properties of Si at 0 K were considered. Several representative reported MEAM potentials [8,9,12,13] were cited as comparison during the verification of the physical properties of Si. Only reported properties calculated by Lee [8], Ryu [9] and Liu [13] potentials were cited, and the calculation wasn't conducted based on these potentials as they are not available for the authors. The properties by Jelinek potential [12] were calculated in the present paper as the potential is available.

4.1. Potential energy

Fig. 1 shows the relation between the potential energy of diamond Si and the first-nearest neighbor distance of Si-Si atoms at 0 K. The potential energy curve given by Rose equation when d_{rep} and d_{att} are zero is also shown in Fig. 1, and it could be seen that it deviates from the DFT value [6], which indicates that the potential has higher accuracy when d_{rep} and d_{att} are not zero. The parameters d_{rep} and d_{att} are adjustable during the fitting process, so the potential energy curve calculated by the present MEAM potential is closer to the DFT value [6], when comparing with the potential energy curves given by Ryu [9] and Jelinek [12] potential.

4.2. Cohesive energies and lattice constants

Table 2 lists the cohesive energies E_c and the lattice constants a of different structures of Si at 0 K. It could be seen that diamond Si is the most stable structure at 0 K. The cohesive energies of different structures of Si calculated by the present MEAM potential agree well with the DFT values [22]. Similar to the other potentials, the lattice constants of the simple cubic, bcc and fcc Si calculated by the present MEAM potential are higher than the DFT values [22]. It should be