Development of Potential Model of Tungsten for Classical Molecular Dynamics Simulation

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1. Introduction

Tungsten (W) is considered as one of the candidate materials for plasma facing components in fusion reactors, thanks to its superior material properties such as low tritium inventory, low sputtering rate, low thermal expansion, and high melting point.

Molecular dynamics (MD) simulation can be used in understanding the behavior of tungsten during the reactor operation. There are two types of MD simulation methods, classical MD (CMD) and first-principles MD (FPMD), which are different in the way they calculate the potential energy and forces needed to simulate time evolution of a system. Although FPMD has a rigid physical background on quantum mechanics (QM), its calculation cost usually scales as $O(N^2)$ or $O(N^3)$, where N is the number of atoms used in simulation, due to its inherent calculation procedures. Therefore, even nowadays, FPMD simulations are limited to around several hundred atoms. In contrast to FPMD, CMD requires relatively low time cost and the time cost is basically linear to the number of atoms, namely $O(N)$. However, for CMD, the preparation of an accurate potential model, which describes interatomic interactions approximately, is a difficult task, and the use of an inaccurate potential model significantly decreases the accuracy of simulation results.

In the present study, we propose a method to systematically construct an accurate potential model for CMD simulation by using calculation results of FPMD. As a validation test of the method, a potential model for tungsten is constructed.

2. Methods

2.1 Potential Model Construction

A potential model is a function to describe the interactions of atoms for energy and force calculations used in CMD. Input parameters of a potential model are characteristic information of atomic position. Because positional configuration of N atom system is perfectly described by $3N$ coordinates of atoms, the potential energy of N atoms can be also perfectly described by $3N$ parameters.

However, to make a potential model practically useful, one needs to reduce the calculation cost and needs to satisfy physical constraints such as rotational and translational invariance. Generally, the interaction over N atoms, which can be regarded as N-atom interaction, is divided into the summation of n-atom interaction ($n < N$). Then, a mathematical formula, which is a potential model, with a small number of parameters for n-atom interactions is constructed such that the physical properties of a target system are reasonably reproduced.

In the present study, we explicitly treat up to three-body interaction and approximately treat many-body interactions above four-body with an embedded-atom method (EAM) potential [1]. As the basis functions of each potential term, namely two body, three body and EAM, we use a Fourier sine and cosine series. The coefficient of each basis function is linearly optimized by minimizing the least square error from a set of energy, force and stress data calculated by FPMD simulations.

The VASP code is used for the FPMD simulations based on the density functional theory (DFT). The Perdew-Burke-Emzerhof (PBE) exchange correlation functional was utilized. Energy, force and stress data are collected in perfect crystals, liquids, defective systems containing a vacancy, a vacancy cluster or a self-interstitial atom (SIA), diatomic molecules and surfaces in a wide temperature range (0 ~ 5700 K).

2.2 Physical Quantities Reproduction

After construction of the potential model, physical quantities are calculated by CMD using the LAMMPS code. Subsequently, the quality of the potential model is evaluated by comparing the calculation results with experimental data and first principles calculation results. The equilibrium lattice constant, cohesive energy, bulk modulus, vacancy formation energy, and the melting point are examined. Experimental data and first principles calculation results are taken from [2], and the melting point is from [3].

3. Results

3.1 Material Properties

The equilibrium lattice constant and the bulk modulus are calculated from a relation between the potential energy and lattice constant of bcc-W crystal, as shown in Fig. 1. The equilibrium lattice constant corresponds to the value at the potential minimum, and the bulk modulus to the derivative of the curve at the potential minimum. Note that the equilibrium lattice constant and bulk modulus calculated in this way are equivalent with the values at 0 K without including zero-point vibration effects. The developed potential model calculates the equilibrium lattice constant to be 3.172 Å, which is in

reasonable agreement with the experimental data and FPMD results [2], about 3.13 – 3.18 Å. The cohesive energy can be evaluated as the difference of tungsten atom energy in a perfect crystal and that in a vacuum. The calculation result is 8.44 eV/atom, which is also comparable with a first principles calculation result (9.97 eV) and an experimental result (8.90 eV) [2]. The bulk modulus is calculated to be 327 GPa, which is comparable with an experimental data (310 GPa) and a first principles calculation result (313 GPa) [2]. The vacancy formation energy is evaluated as 3.47 eV. The experiments data and first principle results were previously reported in a range of 3.15 – 4.6 eV [2].

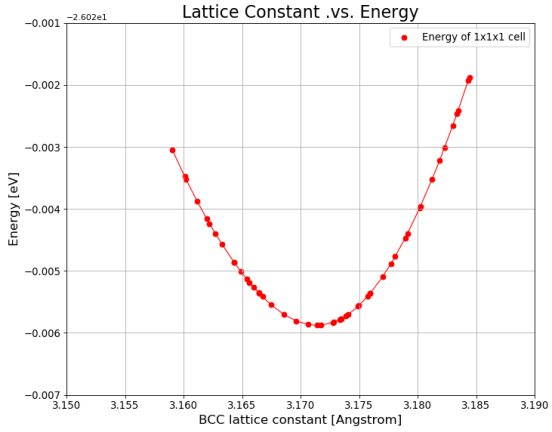
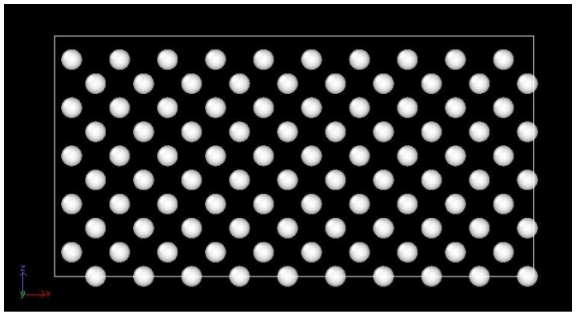


Fig 1. Potential energy calculated by the developed potential model as a function of lattice constant of bcc-W crystal.

The melting point is evaluated by checking the stability of an interface between a solid phase and a liquid phase at several temperatures. In a CMD simulation at 3400 K, the interface disappears and the whole system becomes a crystal, indicating that 3400 K is below the melting point. On the other hand, a simulation at 3600 K results in a liquid phase, indicating that 3600 K is above the melting point. From these results, the melting point is considered to lie between 3400 K and 3600 K (Fig. 2). The experimental melting temperature is 3695 K, while a lower value, 3450 K, has been reported in a first principles calculation [3]. Since the present potential model is fitted to FPMD results, the slight underestimation would be reasonable.



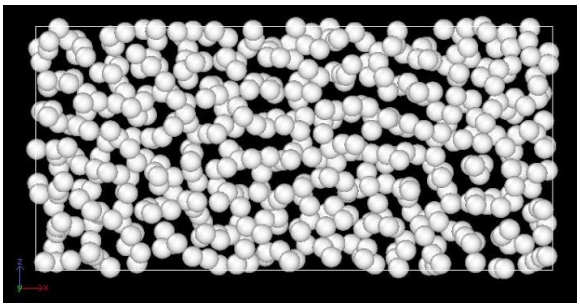


Fig 2. Stable phases at 3400 K and 3600K.
(up: 3400K, down: 3600 K)

3.2 Surface reconstruction

It is known that tungsten shows <100> surface reconstruction [4]. This surface reconstruction is reproduced by first principles calculation [5], while existing potential models fail to reproduce this [6], [7], [8]. In the present study, to reproduce the surface reconstruction, we reinforced the fitting reference, which are energies, forces and stresses calculated by first principles calculation, repeatedly. Specifically, for surface structures that the potential model overestimates the stability, we performed first principles calculation and added the calculation results into the fitting reference. Fig. 3 shows that the energy of the most stable structure obtained by the potential model gradually converges to the energy of the reference reconstructed structure obtained by first principles calculation.

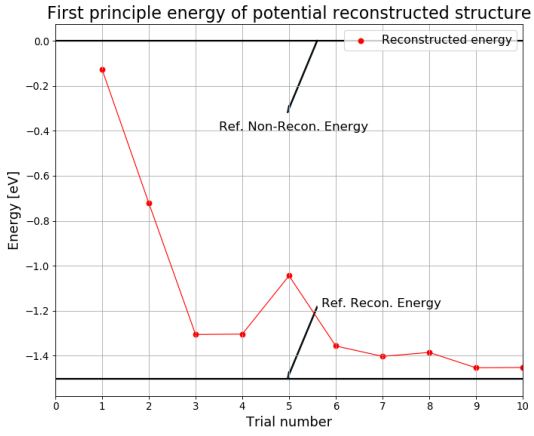


Fig 3. Improvements of potential model in prediction of <100> surface reconstruction.

Table 1 summarizes the quality of the potential model. As can be checked from the table, potential model reproduces the physical quantities in accuracy of the variance of reference data [2][3].

Table 1. Summary of calculated physical quantity in comparison with first principles calculation and experiment.

	Lat. const. [Å]	E_{coh} [eV]	E_{vac} [eV]
Potential Model	3.17	8.44	3.47
QM Calculation	3.13 – 3.18	8.40 – 11.81	3.31 – 3.54

	Bulk Mod. [GPa]	Melting point [K]	Surf. Recon. Ene. [eV]
Potential Model	346	3400 – 3600	−1.4526
QM Calculation	310 – 344	3450 – 3695	-1.5035

3.3 Calculation cost

Finally, the calculation cost of developed potential model is compared with the costs of other existing potential models for tungsten. In this comparison, two types of existing potential models are used: namely, a bond-order potential [6] and an embedded-atom method (EAM) potential [7]. In the bond-order potential, angle dependence is explicitly included, while the EAM potential does not include it. The speed of the three potential models are compared for a CMD simulation of bcc-W crystal, as shown in Fig. 4.

Fig. 4 shows that the EAM potential [7] is the fastest. This is because the angle dependence is not explicitly treated in EAM potentials, different from bond-order potentials and the present potential model. Compared with the bond-order potential [6], the speed of the developed potential model is around one-fourth. This difference in the speed comes from the interpolation scheme that is used for three body terms in the present potential model, which we plan to improve in a future work.

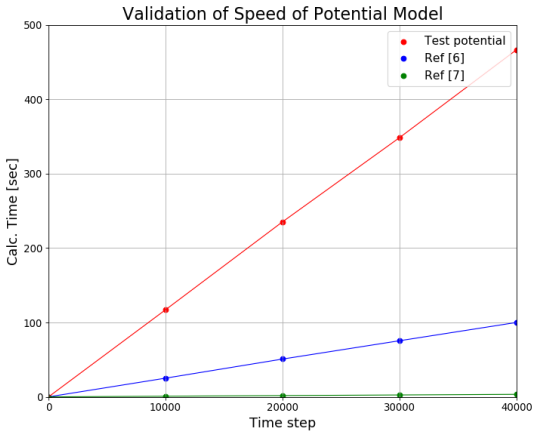


Fig. 4. Comparison of calculation cost of the developed potential model and two existing potential models [6], [7].

4. Conclusions

We have developed a potential model fully based on results of first principles calculations. The developed potential model shows good performance for material properties calculation as well as for simulation of surface reconstruction, which has not been reproduced by other existing potential models. Using this potential model, for example, defect evolution in tungsten under high radiation conditions will be simulated in a future work. Furthermore, by including other first principles calculation results into the fitting reference, such as data on hydrogen or helium in tungsten, simulations of material behaviors under fusion relevant conditions will become feasible, which is expected to provide insights into plasma-surface interactions.

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