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A comparison of interatomic potentials for modeling tungsten nanocluster structures

Jiannan Hao, Xiaolin Shu *, Shuo Jin, Xuesong Zhang, Ying Zhang, Guang-Hong Lu

School of Physics and Nuclear Energy Engineering, Beihang University, Beijing 100191, China
Beijing Key Laboratory of Advanced Nuclear Materials and Physics, Beihang University, Beijing 100191, China

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ABSTRACT

Molecular dynamic simulation is utilized to study the nanocluster and the fuzz structure on the PFM surface of tungsten. The polyhedral and linear cluster structures based on the icosahedron, cuboctahedron and rhombic dodecahedron are built up. Three interatomic potentials are used in calculating the relationship between the cluster energy and the number of atoms. The results are compared with first-principles calculation to show each potential's best application scale. Furthermore, the transition between the icosahedral and the cuboctahedral clusters is observed in molecular dynamic simulation at different temperatures, which follows a critical curve for different numbers of atoms. The linear structures are proved to be stable at experimental temperatures by thermodynamics. The work presents a selection of interatomic potentials in simulating tungsten cluster systems and helps researchers understand the growth and evolution laws of clusters and the fuzz-like structure formation process in fusion devices.

1. Introduction

Tungsten (W) is currently one of the most likely candidates for plasma facing materials of the nuclear fusion devices due to its high melting point, high thermal conductivity, and resistance to ion irradiation properties [1]. However, the dusts sputtering [2–4] from the tungsten surface and hydrogen/helium bubble bursting and fuzz-like structure formation [5–9] can it make hard to operate the plasma reaction [10,11]. Thus, a knowledge of the tungsten nanocluster structures and their properties will help to understand the production and formation processes of the dust and the fuzz structures.

Although first-principles (FP) and molecular dynamics (MD) studies [12–22] of noble metal clusters have been carried out over recent years, few works exist on transition metals especially tungsten [15]. Therefore, the MD method has been used to simulate large systems including hundreds to thousands atoms. The MD simulations are strongly dependent on the interatomic potential.

In this paper, the tungsten clusters in typical polyhedron structures are built up by using the generalized Wulff construction principle [23]. This is one of a number of methods to construct and obtain a stable nanocluster for metals [20–22]. This work aims to compare the different performance of interatomic potentials. Therefore, by using the MD method with three interatomic potentials developed by Juslin [24], Li [25] and Marinica [26], the energy, structure and thermal stability difference are compared. The transition process between two regular structures is observed. First principles calculations have been undertaken for a few representative clusters. Moreover, a few linear structures are built up, and their rationality and stability are verified by the MD and FP calculations. The polyhedral and the linear structure W nanoclusters are the keys to exploring the reasons for the complex surface morphology and for analyzing the plasma irradiation process of tungsten.

2. Methods and models

2.1. Computational methods

Our first-principles calculation were performed using a pseudopotential plane-wave method and generalized gradient approximation (GGA) [27] implemented in the Vienna Ab-initio Simulation Package (VASP) code [28–30]. Through a series of tests, the cutoff energy was set to be 350 eV and a $2 \times 2 \times 2$ k-points by Monkhorst–Pack scheme [31] was used. A large supercell with vacuum space longer than 12 Å containing less than 150 atoms has been utilized. All three cartesian dimensions had periodic boundary conditions. The supercell size was fixed at a constant value with the atomic positions relaxed. The iteration concluded when

* Corresponding author at: School of Physics and Nuclear Energy Engineering, Beihang University, Beijing 100191, China.
E-mail address: shuxlin@buaa.edu.cn (X. Shu).

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forces on all atoms became less than 10^{-3} eV/Å. All the atoms were initialized as charge neutral and the electric magnetic moment was not considered for tungsten, acknowledged as a paramagnetic metal.

The MD LAMMPS code is used to study the nanocluster structures of tungsten. The simulation box volume had a length of 300 Å to keep a vacuum space between clusters, so avoiding interactions. Then, three tungsten potentials are compared: A Finnis-Sinclair potential [32] for W-He modified at short range by Ackland and Thetford [33] and by Juslin and Wirth [24]; a Tersoff-type [34] bond-order potential (BOP) for W-H-He developed by Li et al. [25]; an embedded atom model [35] (EAM) potential for W defects by Marinica et al. [26]. All three potentials were used only in simulations of the W-W interaction part.

For comparison, a few basic properties are shown in Table 1. The BCC lattice constant α, cohesive energy (E_c) and surface energy were calculated in this work using three potentials and the VASP code. The Δγ represents the surface energy difference between γ (1 0 0) and γ (1 1 0). Fundamental properties for bulk and surface of the three interatomic potentials are mainly fitted to the experimental data and first-principles calculations.

2.2. Models

All polyhedral nanoclusters simulated in this work were built following geometry rules for icosahedral (ICO), cuboctahedral (CUB) and rhombic dodecahedral (RHO) clusters seen in Fig. 1 (a)–(c). The ICO is one of the five regular Platonic solids, and is represented by its Schläfli symbol {3, 5}, containing 20 triangular faces, with 5 faces meeting around each vertex. Then the CUB is a polyhedron with 8 triangular faces and 6 square faces, which has 12 identical vertices, with 2 triangles and 2 squares meeting at each vertex, and 24 identical edges, each separating a triangle from a square. In this work, the CUB cluster is compressed via a normal vector of a square face due to the BCC atomic interaction, whose energy distribution is discussed in Fig. 2. Moreover, from geometry, the RHO is a convex polyhedron with 12 congruent rhombic faces, which has 24 edges, and 14 vertices of two types. It is noted that W with the RHO structure has 12 faces oriented all six close-packed planes of BCC metals. The atoms shown in Fig. 1 are displayed in a rainbow color, where the potential energy of red atoms is higher than that of blue ones. Fig. 1(d) shows a spherical cluster compared to polyhedral structures, which apparently has more red atoms than other polyhedra. Though the spherical structures seem to be the most symmetric shape at the macro level, it has more atoms with high energy than the polyhedron and may be unstable when different potentials are used. Therefore, we built those polyhedral structures in order to obtain relative comparable results.

In addition, there are two linear structures abbreviated to C5 and C6 studied in this work respectively. These are the structures seen in Fig. 1(e), (f) with a cross-section drawing seen in Fig. 6. Each segment of C5 belongs to a five-fold cyclic group meaning it overlapped itself after 72° rotation along the Z-axis. The C6 structures is similar to C5 with a six-fold symmetry. All these atoms are free of strain and placed in a vacuum environment without any restraints, besides the periodic boundary conditions used to calculate the infinite linear structures in MD simulations.

3. Results and discussion

3.1. Polyhedral clusters

The average potential energy for the three polyhedral nanoclusters as the function of the number of atoms in MD and FP simulations are shown in Fig. 3. The ICO structure with a high symmetric regular polyhedron has the lowest potential energy among the three structures by MD simulations with the three potentials when the number of atoms is 13 or 55, which is shown in Fig. 3(a)–(c). However, the RHO structure has the lowest average energy when the number of atoms is over 147 by Marinica's and Li's potentials, and over 309 by Juslin's potential. Because the RHO structure has 12 faces oriented to the W(1 1 0) close-packed planes, this has the lowest surface energy for W, while the ICO inner atoms are ordered in FCC and HCP mixed configuration which is not stable for BCC W. These are two advantages to make the RHO structure emerge as the cluster size increased. Then, the DFT result shows that RHO always has the lowest average energy in our calculations. The MD and FP results imply these three potentials may perform differently when reproducing extremely small nanocluster behaviors.

Though ICO has a higher energy in FP calculations for W metal without thermal dynamics considered, it shows stable high symmetric and magic sized structures in other FCC or HCP metals [13,15,16,20]. Zhao's work [20] indicated that the edge atoms play an important role in the formation of nanoclusters, and the ICO structure has the lowest energy among other structures with the same number of atoms. While Lin [36] gives the W nanocluster ground-state with 13 atoms not in the ICO structure, which supports our calculations. For instance, the potential energy for each atom of the CUB structure with 561 atoms, which is convenient for comparing energy difference, is displayed in Fig. 2 and the structure shown in Fig. 1(b). The dash line represents the cohesive energy calculated by potentials, which indicates that the energy of the sub-surface is similar to that of the bulk. The points between solid lines represent the same layers. Then, the layers are separated into three parts by dot lines for faces, edges and vertices. The energy difference of (1 0 0) and (1 1 0) faces are consistent with the calculations in Table 1.

In addition, we also found a mutual change between the ICO and CUB structure during dynamic simulation from 0 K to 3000 K, because these two structures share the same numbers of atoms in each shell with both 12 vertices. The black line in Fig. 4 shows the critical transition temperature borderline between the CUB and ICO structures. The CUB structure can exists at a temperature below the borderline. But the CUB structure will transform to the ICO structure as the temperature rises up over the borderline. However, the ICO structure could not transform to CUB by a temperature reduction. What is more, the potential energy of ICO is much close to that of CUB when the atom number is 309 calculated

Table 1
The fundamental properties of bulk and surface using the three interatomic potentials compared to experiments and first-principles results.

	Experimental	First-principles	Juslin	Li	Marinica
α (BCC) (Å)	3.165 [41,42]	3.174	3.165	3.165	3.140
Ec (BCC) (eV)	−8.90 [41]	−8.59	−8.90	−8.91	−8.90
γ(1 1 0) (J/m^2)	3.265 [43] 3.675 [44]	4.005 [45]	2.576	2.319	2.306
γ(1 0 0) (J/m^2)		4.635 [45]	2.924	3.157	2.721
Δγ (J/m^2)	–	0.630	0.348	0.838	0.415

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