Nuclear Instruments and Methods in Physics Research B 410 (2017) 171–178

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

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

A molecular dynamics study of coalescence of tungsten nanoparticles

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ARTICLE INFO

Article history:
Received 10 May 2017
Received in revised form 21 August 2017
Accepted 21 August 2017

Keywords:
Molecular dynamics simulations
Tungsten nanoparticle
Coalescence

ABSTRACT

The coalescence behavior of tungsten nanoparticles was investigated using molecular dynamics simulations. Two free tungsten nanoparticles and tungsten nanoparticles on tungsten (0 0 1) substrate were studied. For free tungsten nanoparticles, the nanoparticles size, temperature and relative velocity were found to affect the coalescence. The surface effect had a considerable influence during the coalescence process. For coalescence of two tungsten nanoparticles on tungsten (0 0 1) surface, considerable deformation of nanoparticles and significant epitaxy on the nanoparticle-surface interface were observed. When the aligned structures were bombarded with other nanoparticles, the change in surface morphology was found to be more complicated.

1. Introduction

In recent years, metal nanoparticles ranging from 1 to 100 nm have been studied extensively, both experimentally and theoretically [1–8], because they are a bridge between the bulk materials and atomic and molecular structures. Considerable efforts were made to investigate the basic thermal structure and melting behavior of metal nanoparticles [1,4,5,9–12]. The interaction between metal nanoparticles and surface has also attracted much attention [9,13–16].

Because of its high melting point, low sputtering yield, high density and hardness, tungsten has been selected as the plasma-facing material (PFM) [17] in controlled nuclear fusion reactors. Tungsten dust was found in fusion devices, including those on a nanometer length scale. Some tungsten dust deposited on PFM is coalesced with multi-nanoparticles [18]. The coalescence behavior may occur during the flight process of dust or after the deposition on PFM. The process is difficult to be investigated in plasma in experiments directly [19]. The essential structure change cannot be investigated in detail either. Thus, theoretical studies such as molecular dynamics (MD) simulations are needed.

For free nanoparticles, Moitra et al. [20,21] presented an MD study of melting and sintering behavior of tungsten nanoparticles. The study showed the process of the coalescence of tungsten nanoparticles directly. However, the initial temperature of two nanoparticles were the same, and there was no further investigation of the influence of initial size and configuration of nanoparticles on sintering. In fusion devices, free nanoparticles move with different velocities. They may be accelerated by magnetic fields and gain more kinetic energy. When they collide with each other, the result is more complicated.

For nanoparticles deposited on tungsten substrate, Luo et al. [22] studied the deposition of tungsten nanodust on tungsten surface at 100 m/s impinging speed with MD, and proved that the dust adhered to the wall with slight deformation. The loosely bounded dusts may coalesce into a larger dust easily. The cluster soft landing has been studied with much effort during past years [9,13,15,23,24]. Epitaxy is easy to be observed after cluster soft landing. The stable epitaxy may be an obstacle to the coalescence of nanoparticles, which need to be investigated. What will happen to the coalesced structure when another nanoparticle bombards with impacting energy is uncertain.

We successfully studied the melting and dissociation of free nanoparticles with diameters ranging from 4 to 12 nm with MD [12]. In this work, we focused on tungsten nanoparticles with diameters below 6 nm and studied the coalescence between them. The coalescence of free nanoparticles (during the flight) and nanoparticles on tungsten substrate (after deposition on PFM) was investigated.

2. Method

An MD package Molecular Dynamics Package of Sichuan University (MDPSCU) with graphics processing units (GPUs) for parallel computing was used [25].

To study the coalescence of the tungsten nanoparticles with different initial states, we needed to investigate the melting

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http://dx.doi.org/10.1016/j.nimb.2017.08.031
0168-583X/

temperature of nanoparticles. We first generated a cubic tungsten box at 0 K. Tungsten nanoparticles with different diameters were cut from the box. Then, the temperature of each nanoparticle was slowly increased from 0 K by adopting an electron-phonon coupling model proposed by Finnis et al. [26], which has been used for simulating the depositions of Au clusters on surfaces [13] and the bombardments of He atoms on tungsten surfaces [27]. A large characteristic time of 500 ps was applied to make sure the nanoparticle was in quasi-equilibrium. The process lasted 1,000,000 time steps (1 ns), and the final temperature of the nanoparticle was higher than 6000 K. The surface boundary condition was free, and the lattice length a_0 was set as 3.1652 Å. During the process, the evolution of the cohesive energy (C.E.) per atom in each nanoparticle with increasing temperature was obtained. A Finnis-Sinclair-type potential obtained by Ackland et al. (ACK) [28] was adopted to describe the interaction between tungsten atoms. The detailed information of the melting temperature is presented in Ref. [12].

To study the coalescence of free tungsten nanoparticles, the influence of nanoparticle size, temperature and relative velocity was considered. First, the coalescences of 2 nm (272 atoms) −2 nm nanoparticles and 6 nm (7192 atoms) −6 nm nanoparticles were simulated and compared to study the influence of particle size on coalescence. The temperature was set as 2000 K. Then, for the influence of temperature, three cases were considered: coalescence of two solids (2000 K–2000 K), of two liquids (4000 K–4000 K), and of a solid and a liquid (2000 K–4000 K, 2000 K–6000 K). All the nanoparticles were prepared with 6 nm diameters. Finally, to study the effect of relative velocity on coalescence, velocity was added to one 6-nm nanoparticle and made to collide with another static one. The temperature was set as 2000 K. Different relative velocities in the range of 100 m/s–3000 m/s were considered. The distance between two nanoparticles was set as one lattice length initially. The processes lasted 1000 ps, and the size of the time step was adapted to guarantee the maximum distance for the atoms moving in one time step in less than a given threshold (0.08 a_0). The surface boundary condition was free. To investigate the effect of nanoparticle size, temperature and relative velocity on coalescence more precisely, two nanoparticles in each coalescence case were set with same axial directions and their mass centers were along the same ⟨0 0 1⟩ direction.

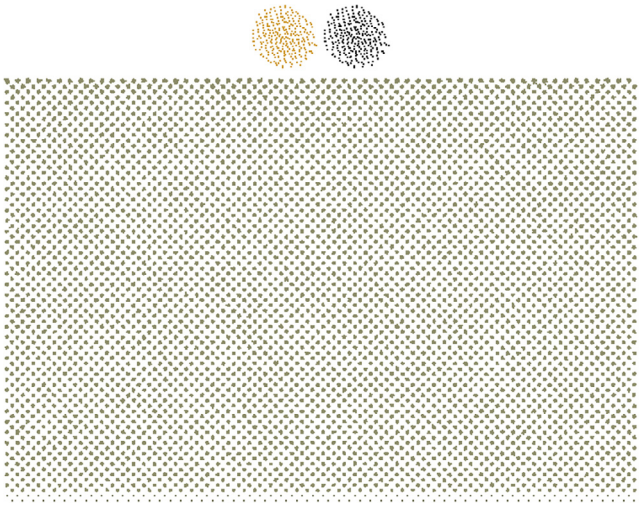

Fig. 1. Simulation model for coalescence of two 2-nm nanoparticles on a tungsten (0 0 1) substrate. Yellow and black dots: nanoparticle atoms; gray dots: substrate atoms. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

To study the coalescence of tungsten nanoparticles on tungsten substrate, a tungsten (0 0 1) box was thermalized and relaxed to thermal equilibrium at 2000 K. The size of the box was 60 a_0 - × 60 a_0 × 40 a_0, containing 288,000 atoms. Two layers at the bottom were fixed to avoid net drift of the atoms in a vertical direction. Two nanoparticles (2 nm–2 nm, 6 nm–6 nm, 2000 K) were placed above the tungsten surface with one lattice length distance. Fig. 1 shows an example of two 2-nm nanoparticles placed on a tungsten (0 0 1) substrate. Periodic boundary conditions were applied in x- and y-directions. The process lasted at least 100 ps, and the interactions between nanoparticles and between nanoparticles and the tungsten surface were investigated. The size of the time step was set the same as above.

When the system mentioned above was stable, another 2-nm nanoparticle at 4000 K was placed above the surface again, and collided with the deposited nanoparticles with a velocity ranging from 1000 m/s to 8000 m/s. The simulation lasted 100–1000 ps, depending on the velocity. During the process, the changes to the nanoparticles and the tungsten surface were studied.

3. Results and discussion

3.1. Coalescence of free tungsten nanoparticles

3.1.1. Effect of nanoparticle size on coalescence

To study the influence of particle size on coalescence, different diameters were considered. We first present the coalescence of two 6-nm nanoparticles at 2000 K. The tungsten nanoparticles are now in a solid state. The coalescence process is shown in Fig. 2(a–c). Two nanoparticles are represented with different colors. The process is the same as in Ref. [21]. A "neck" appears between two clusters (Fig. 2(b)), making the system seems like a dumbbell. However, the "neck" lasted a long time. Even at 10 ns, very few atoms diffused away from the contact region (Fig. 2(c)). In Ref. [21], although two 10-nm nanoparticles are also at 2000 K, the "neck" changed much faster. This is because of the application of different interatomic potential.

We presented the coalescence of two smaller nanoparticles with diameters of 2 nm, also at 2000 K. The coalescence process is shown in Fig. 2(d–f). The "neck" after 400 ps was already equal to 2 nm (Fig. 2(e)). The "neck" stayed until 500 ps. Then, a many-faceted nanoparticle with a uniform structure was formed (Fig. 2(f)). Obviously, the two initial solids have a better coalescence with more tungsten atoms diffusing into each other. In fact, at around 1.5 ns, the formation of the many-faceted nanoparticles shown in Fig. 2(f) has already began. Then, the rearrangement and diffusion of atoms made the coalesced nanoparticles more stable and uniform.

Here we define R_d as the distance between the centers of mass of two nanoparticles. The results of two 6-nm solid nanoparticles and two 2-nm solid nanoparticles are shown in Fig. 3. For 6 nm–6 nm coalescence, R_d decreased 5.7 Å in the first 50 ps. Then, R_d stayed around 57.6 Å for a long time, which is in accordance with the snapshots shown in Fig. 2(a–c) and proves the diffusion of atoms is weak. For 2 nm–2 nm coalescence, tungsten atoms diffused faster. Thus, R_d decreased faster than that of 6 nm–6 nm solid nanoparticle coalescence. At 10 ns, R_d already decreased to 4.03 Å.

During the coalescence process, the temperature T of the whole system increased with increase in time t, as shown in Fig. 4. Especially for 2 nm–2 nm coalescence, the average increase in T after 5 ns is about 550 K. This is because the coalescence of two clusters lead to the decrease of the total surface area and the total surface energy. For 2 nm–2 nm coalescence, two clusters merged into one particle with a uniform structure. The total surface area of the systems shown in Fig. 2(d) and (f) is about 31 nm^2 and 24 nm^2, respec-