Ceramics International (xxxx) xxxx–xxxx

Contents lists available at ScienceDirect

Ceramics International

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

Molecular dynamics simulation of AlN thin films under nanoindentation

Henggao Xiang[a], Haitao Li[a,b,*], Tao Fu[a], Yinbo Zhao[a], Cheng Huang[a], Gang Zhang[a], Xianghe Peng[a,b,*]

[a] College of Aerospace Engineering, Chongqing University, Chongqing 400044, China
[b] Chongqing Key Laboratory of Heterogeneous Material Mechanics, Chongqing University, Chongqing 400044, China

ARTICLE INFO

Keywords:
AlN film
Indentation
B4-B1 phase-transition
Amorphization
Dislocation glide
MD simulation

ABSTRACT

A large-scale molecular dynamics (MD) simulation of nanoindentation was performed to study the structural deformation on wurtzite aluminum nitride (B4-AlN). The nanoindentation induced B4-B1 phase-transition, amorphization and dislocation glide in AlN (0001) thin films were found. It shows that the B4-B1 phase-transition path includes two processes: an anti-parallel vertical movement of N and Al atoms along the [0001] axis, followed by horizontal rearrangements of the two types of atoms. Indentation force-depth (*P-h*) curve shows minor and major pop-in events. Detailed analysis of the results shows that the first three minor load drops in the *P-h* curve are related to the nucleation of amorphous structure, whereas the subsequent major load drop is related to the dislocation nucleation and expansion. The dislocations in AlN thin film involve perfect dislocations and Shockley partial dislocations, the latter is associated with the formation of intrinsic stacking faults (SFs) type I_2 during the expansion of dislocation loops.

1. Introduction

Transition metal nitrides have attracted much attention over the past few decades due to their excellent mechanical and physical properties [1–3]. Aluminum nitride (AlN), one kind of metal nitrides, has the characteristics of short bond length, wide band gap, high dielectric strength, low expansion coefficient and large bulk modulus [4,5]. Because of these extraordinary performances, AlN has been widely used not only as piezoelectric films [5,6] but also as negative electrode of lithium battery [7], engineering protective coatings [8] and high-temperature transistors. During applications, AlN film may undergo severe external loading that can cause structural transformation. In addition, AlN has both crystalline and amorphous structures [9]. The crystalline structures include: wurtzite structure (B4) at ambient condition; metastable zinc-blende structure (B3) that can be stabilized under non-equilibrium deposition parameters; rock-salt structure (B1) under high pressures; and intermediate graphite-like (GL) phase, which has been proved to be stable [10]. However, less research progress related to the amorphous structure (AS) of AlN can be found in the literature.

To date, great progress has been made in experimental investigation and first-principles calculations to prove the existence of phase-transition in AlN. For example, Ueno et al. [11] and Xia et al. [12] observed the transformations under high pressure using X-ray. Guo et al. [13] studied the sample size induced the brittle-to-ductile transition in AlN with compression experiment. Based on first-principles calculations, Zhang et al. [14] and Durandurdu et al.[15] studied the B4 to B1 transformation in AlN and Wright et al. [16] investigated the basal-plane SFs in B4-AlN. Nevertheless, because of the limitation of experimental observation on defects and their evolution and the low efficiency of first-principles calculation, some important issues concerning the atomic configuration evolution in AlN film remain unsolved.

The nanostructural properties of materials, such as elastic modulus, amorphization, dislocation and transformation, can be investigated with nanoindentation technique [17–22]. MD simulation may play an important role in predicting the behaviors of nanostructural materials. Branicio et al. [23–25] revealed with MD simulation the mechanism of fracture in AlN induced by hypervelocity projectile impact.

Different from the previous studies, we investigate in this article the nanostructures in AlN (0001) thin films and the behavior of B4-B1 phase-transition path, and analyze in details the amorphization and dislocation glide. Combined with the indentation *P-h* curve, the corresponding deformation mechanisms are also analyzed. In Section 2, the adopted interatomic potential, simulation details and identification technique for local structure are introduced. The simulation results and the corresponding discussion are presented in Section 3. Some conclusions are drawn and given in Section 4.

* Corresponding authors at: College of Aerospace Engineering, Chongqing University, Chongqing 400044, China.
E-mail addresses: htli@cqu.edu.cn (H. Li), xhpeng@cqu.edu.cn (X. Peng).

http://dx.doi.org/10.1016/j.ceramint.2016.11.218
Received 13 November 2016; Accepted 30 November 2016

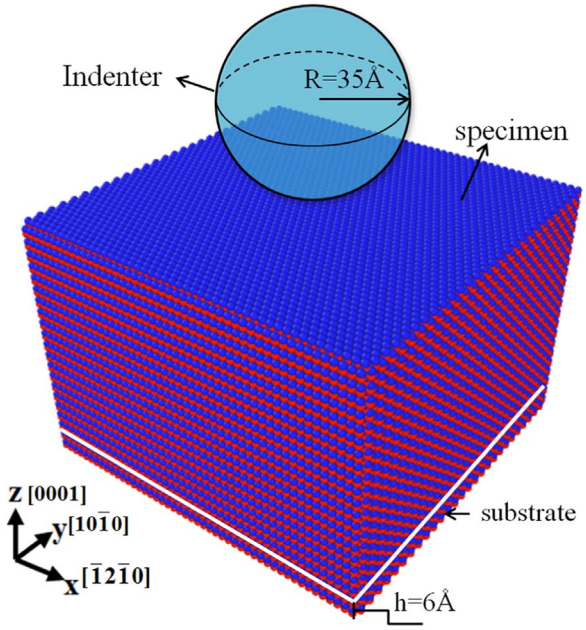

Fig. 1. 3D MD model for AlN under nanoindentation with spherical indenter.

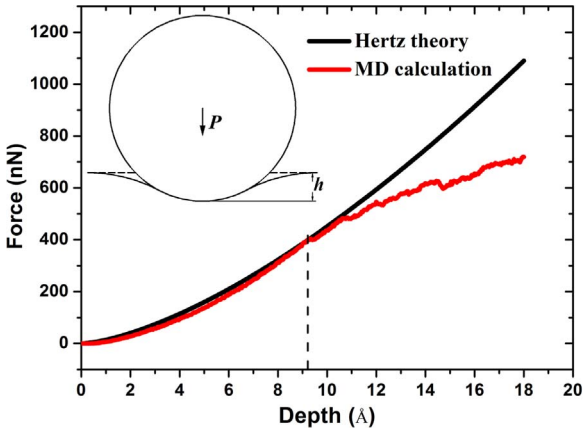

Fig. 2. Comparison between *P-h* curves by MD simulation and by Hertz theory.

2. Methods

2.1. Interatomic potentials

Many interatomic potentials have been developed for AlN [26–30] in the past decade. In this work, the Vashishta potential [31,32], which has successfully been used to describe the behavior of semiconductors and ceramic materials [33,34], is selected to describe interaction in an Al-N system. The main reason for choosing this potential is that it can predict the properties of AlN with various phases. For example, the critical pressure for phase-transition from B4 to B1, calculated with Vashishta potential [29], is 24 GPa, which is consistent with the experimental value of 20 GPa [35].

The indenter is assumed to be a non-atomic repulsive sphere to save computation time. It is repulsive to all the atoms it contacts, with the interaction potential written as [36]

$$V(r) = \begin{cases} k(R-r)^3, & r < R, \\ 0, & r \geq R. \end{cases} \quad (1)$$

where r is the distance between the center of the indenter and that of an atom, R the radius of the indenter and k the stiffness of the indenter, which is set as 10 eV Å^{-3}[37].

2.2. Simulation details

MD simulations for the indentation on AlN (0001) films are performed to investigate the nanostructures. Fig. 1 shows the model for the simulation of the indentation on AlN, where the spherical indenter is R=35 Å in radius. The axes x, y and z are along [$\bar{1}$2$\bar{1}$0], [10$\bar{1}$0] and [0001] orientations of an AlN lattice, respectively. The size of the simulation box is 149.28 Å×145.44 Å ×104.496 Å, which contains 217728 atoms. Making use of the conjugate gradient (CG) algorithm, the specimen is optimized before indentation to achieve a stable configuration with the minimum equilibrium energy. The *NVE* ensemble is chosen in the simulation and the loading rate of nanoindentation is 40 m/s. Langevin thermostat [38] is used to ensure the indentation to be performed at 10 K. The atoms on the bottom of the sample are fixed to prevent the substrate from shifting during indentation. Free boundary condition is set in the upper surface, and periodic boundary conditions in x and y directions, respectively. Before indentation, the indenter is placed 2 Å above the center of the upper surface, and then moves downwards against the substrate. All the simulations are performed using LAMMPS package developed by Plimpton [39] for large-scale three-dimensional (3D) MD simulations.

2.3. Identification of local structure

In order to identify the structural change and defects nucleation and evolution, the methods of coordination number (CN), radial distribution function (RDF) and identify diamond structure (IDS) are used. CN describes the number of neighbor atoms from a central atom within a specified cutoff distance. The distance is set as 2.5 Å in this article, which is determined by the distance between first neighbor and second neighbor. Within the cutoff distance, the perfect B4-AlN phase, B1-AlN phase and intermediate GL phase are of fourfold coordination, sixfold coordination and fivefold coordination, respectively. RDF is normalized by the number density of particles and can be used to identify the lattice structure of crystalline and amorphous [33], and in this work, it will be used to identify the amorphization in deformation zone. IDS is adopted to identify the atoms arranged in hexagonal or cubic diamond lattice. To classify a central atom, the second nearest neighbors are taken into account to distinguish between hexagonal and cubic diamond structures. This method involves the characterization of the geometrical arrangement of the second nearest neighbors and can be used to visualize and identify the stacking faults (SFs) and dislocations [40]. In order to display more clearly the defect region, the local lattice structures are indicated by colors: hexagonal diamond structure (first neighbor) is represented by yellow, hexagonal diamond structure (second neighbor) by dark yellow, cubic diamond structure by blue and the others by green.

3. Results and discussion

3.1. Comparison with Hertz theory

In order to further verify the accuracy of MD simulation, the indentation *P-h* curves of the AlN film, obtained by Hertz theory and by MD calculation, are compared, as shown in Fig. 2. The inset is the diagram for the contact between a rigid sphere and an elastic half space with flat surface. In Hertz theory the indentation force P is related to indentation depth h, indenter radius R and the elastic properties of the contacted materials, as expressed as [41]

$$P = \frac{4}{3}E^* R^{1/2} h^{3/2} \quad (2)$$

where

$$\frac{1}{E^*} = \frac{1-\nu_1^2}{E_1} + \frac{1-\nu_2^2}{E_2} \quad (3)$$