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Atomistic mechanisms for temperature-induced crystallization of amorphous copper based on molecular dynamics simulation

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ABSTRACT

Atomistic mechanisms for temperature-induced crystallization of amorphous copper were investigated by means of molecular dynamics (MD) simulation. During the crystallization, three kinds of atom structures are formed, where face-centered cubic (FCC) structure is dominant, and body-centered cubic (BCC) structure possesses the lowest fraction. The crystallization of amorphous copper is a process of atom diffusion, where atoms in an unstable state are activated by the heat energy and then they diffuse to the locations in which their energy is minimum. During the diffusion, BCC structure is initially formed in the amorphous copper because it possesses a larger volume. As a consequence, both the close-packed hexagonal (HCP) structure and the FCC structure are transformed from the BCC structure. The crystallization of amorphous copper results in plenty of dislocations, which are formed through two approaches. The first approach is the plastic slip induced by the thermal stress, which may be aroused by the thermal-expansion coefficient difference between the crystallized atoms and the amorphous ones. The second approach is the phase transformation, which refers to the transformation from HCP structure to FCC structure in the present study.

1. Introduction

Nanocrystalline materials have attracted a great deal of academic and practical interest in recent years due to their excellent chemical and physical performance as well as mechanical properties [1]. Over the last decades, researchers have developed several methods of fabricating the nanocrystalline materials, including ball milling, severe plastic deformation, inert gas condensation and pulsed electrodeposition [2–5]. Afterward, the method that a nanocrystalline material is fabricated by crystallizing an amorphous material was proposed [6,7].

It is well known that amorphous phase is a thermodynamic instability phase since crystallization can be induced under the action of heat or stress [8,9]. Bulk nanocrystalline materials can be obtained by means of controlling the crystallization conditions. As a consequence, more and more researchers have devoted themselves to investigating the crystallization of amorphous materials by means of experiments. Wang et al. [10] fabricated Co_2MnSi amorphous films by virtue of sputtering at room temperature and they found that the crystallization of the films occurs when they are annealed at 400 °C. Kim et al. [11] studied crystallization behaviors of $Ti_{45}Cu_{40}Ni_{7.5}Zr_4Sn_{2.5}Si_1$ amorphous-based composite with B2 particles via isothermal heat treatment, and they found that two steps occur during the crystallization of the amorphous phase, where the nanocrystalline phase is formed firstly and subsequently γ-TiCu phase is precipitated. Wu et al. [12] investigated the crystallization behavior of $Fe_{78}Si_{13}B_9$ amorphous alloy induced by Ar ion bombardment, and they found that the crystallization in this case is a phase transformation process induced by stress as well as thermal effect. Svoboda and Málek [13] studied crystallization kinetics of a Se-Te glassy system, which is composed of a set of powder fractions divided by particle size, by means of non-isothermal treatment, and they proposed that the crystallization is initiated from the nuclei randomly distributed in each glassy particle, the defects induced by grinding and milling, and the stress-induced defects. Zhan and Ernst [14] investigated the micro-mechanism for the crystallization of near-eutectic amorphous Ni-P by virtue of isothermal tempering, and they found that the microstructure of crystallizing phases is similar to what occurs during the solidification of a eutectic melt. Kozyukhin et al. [15] studied the crystallization of deposited $Ge_2Sb_2Te_5$ amorphous thin films by menas of isothermal annealing and continuous-wave laser irradiation, and they suggested that the crystallization was induced by the heterogeneous nucleation on the film boundary. Park et al. [16] investigated the influence of residual thermal stress on the crystallization of hydrogenated amorphous silicon films via annealing and capping the SiN_x and Al_2O_3 layers, and the results indicate that tensile stress is able to enhance the crystallinity of the annealed amorphous silicon film. Bakkal et al. [17] found crystallization phenomenon during machining

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a Zr-based bulk metallic glass. Even though the aforementioned investigations have found the laws for the crystallization of some amorphous materials in a certain extent, they are unable to reveal the fundamental mechanisms because the evolution of the crystallization process is invisible via conventional experiments.

As a powerful tool for studying the physical mechanisms for plastic deformation and crystallization of materials at atomistic scale, molecular dynamics (MD) method has been widely used in material researches [18–24]. However, only a few literatures related to crystallization of amorphous materials have been found. Lee et al. [25,26] investigated the deformation behavior of amorphous Ni thin films under tensile stress as well as hydrostatic pressure via MD simulations, and they found that the stress plays an important role in the crystallization of amorphous Ni thin films. Park et al. [27] studied the isothermal crystallization induced by externally cyclic forces via MD simulation, and they found that the enhanced force field is able to effectively induce the crystallization without heating action. Yang et al. [28] studied the heat-induced crystallization of amorphous Ni_xZr_{100-x} (x = 10, 16.7 and 33.3) alloys by virtue of MD simulation, and they found that the crystallization of amorphous alloys during heating is related to the alloy composition and the heating rate. Timonova and Thijsse [29] investigated the crystallization process of amorphous Si under high-temperature treatments via MD simulations, where crystallization dynamics was discussed. Zhao et al. [30] investigated the stress-induced crystallization of amorphous copper in the course of cutting via MD simulation, and they found that the crystallines are nucleated in the larger-stress regions on the rake face and the tip of cutting tool and they subsequently grow quickly under the stress.

Even though the physical images of crystallization process have been captured in the aforementioned MD simulations, the mechanisms of the crystallization have not been clearly revealed. As a consequence, MD simulation was applied in the present study so as to investigate the atomistic mechanisms for the temperature-induced crystallization of amorphous copper, where both the mechanism of structure evolution and the one of crystallization dislocations were revealed.

2. Modeling and method

2.1. Simulation models

In the present study, the amorphous copper model used to simulate the crystallization was established by liquid melt quenching. The initial crystalline model with the size of 30 × 30 × 30 unit cells (a = 0.3615 nm) was generated as a face-centered cubic (FCC) lattice, where the [1 0 0], [0 1 0] and [0 0 1] directions of the crystal are parallel to x, y and z directions of Cartesian coordinate system, respectively. Firstly, the initial FCC model was relaxed for 5 ps at 300 K under the NVT ensemble. Subsequently, the model was heated to 2000 K at a constant heating rate of 1.7×10^{14} K/s and then was cooled to 300 K at a constant cooling rate of 8.5×10^{15} K/s. The model was equilibrated for 100 ps at 2000 K so that the complete liquid phase was obtained at the end of the equilibrium time. Both heating and cooling processes were performed under the NPT ensemble. As a consequence, an amporphous model was obtained by the above fast quenching method. Fig. 1(a) and (b) show the initial FCC model and the quenched amorphous model, respectively. In the course of crystallization simulation, the system was heated from 300 to 900 K at a constant heating rate of 2.5×10^{13} K/s and then was held for 200 ps at 900 K. Periodic boundary conditions were adopted in the three dimensions of the simulation box during both amorphization and crystallization courses.

2.2. Interatomic potentials

The embedded-atom-method (EAM) potential developed by Minish et al. [31] was applied in the current investigation for the purpose of describing the interactions among copper atoms in the system. This potential was used to predict reliably energies and stability of nonequilibrium structures by taking Cu as a model material, so all the potential parameters in the present study were the same as those in this potential. In the EAM potential, the total energy of a system is described as

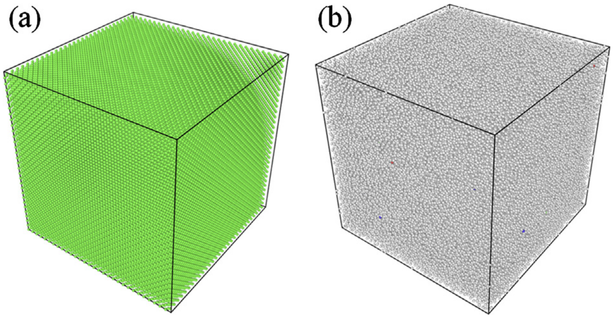

Fig. 1. Simulation model: (a) Initial FCC model; (b) Quenched amorphous model.

$$E_{\text{tot}} = \frac{1}{2}\sum_{ij} V_{ij}(r_{ij}) + \sum_{i} F_i(\bar{\rho}_i) \tag{1}$$

where V_{ij} is the pair potential between atoms i and j, r_{ij} is the distance, F_i is the embedding energy of atom i, and $\bar{\rho}_i$ is the host electron density at the site of atom i. $\bar{\rho}_i$ is induced by all the other atoms in the system and it is calculated by

$$\bar{\rho}_i = \sum_{j\neq i} \rho_{ij}(r_{ij}) \tag{2}$$

where ρ_{ij} is the electron density as a function of r_{ij}.

2.3. Simulation method

The MD simulation was conducted by virtue of LAMMPS code developed by Plimpton [32]. For the purpose of verifying the crystallization evidence, radial distribution function (RDF) was adopted to monitor atom distributions. The RDF, which is expressed by $g(r)$, is a fundamental function used to describe the liquid and disordered systems. For a given atom i, RDF reflects the probability of finding another atom in a space ranging from r to r + dr, where r is the distance from atom i to other atoms. In addition, the RDF describes the distance distribution between an atom and other atoms around it. For an ideal lattice structure, the RDF presents peak values in different neighborhoods. The disappearance of key peaks demonstrates that the melting or the disorder occurs in the system. According to the definition, the RDF is expressed as [33].

$$g(r) = \frac{n(r)}{\rho_0 V} \approx \frac{n(r)}{4\pi r^2 \rho_0 \mathrm{d}r} \tag{3}$$

where $n(r)$ is the number of atoms and V is the corresponding volume; ρ_0 is the atomic density of a perfect crystal, and it is equal to 4 for a FCC structure; r is the distance from the distributed atoms to a given atom.

In order to provide information for diffusion behavior of atoms, mean square distance (MSD) calculation was performed in the present study. According to the definition, MSD can be mathematically expressed as [34]

$$r^2(t) = \frac{1}{N}\left\{\sum_{i=1}^{N} |\vec{r_i}(t) - \vec{r_i}(0)|^2\right\} \tag{4}$$

where N is the atom number in the system, and $\vec{r_i}(t)$ is the position of atom i at time t.

The visual software OVITO [35] was used to investigate the atom distribution and dislocation evolution during the crystallization. In order to identify the crystal structure of the atoms, common neighbour