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Molecular dynamics simulations of aggregation of copper nanoparticles with different heating rates

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ABSTRACT

Molecular dynamics simulations were employed to investigate the heating rates' effect on aggregation of two copper nanoparticles. The aggregation can be distinguished into three distinct regimes by the contacting and melting of nanoparticles. The nanoparticles contacting at a lower temperature during the sintering with lower heating rate, meanwhile, some temporary stacking fault exists at the contacting neck. The aggregation properties of the system, i.e. neck diameter, shrinkage ratio, potential energy, mean square displacement (MSD) and relative gyration radius, experience drastic changes due to the free surface annihilation. After the nanoparticles coalesced for a stable period, the shrinkage ratio, MSD, relative gyration radius and neck diameter of the system are dramatically changed during the melting process. It is shown that the shrinkage ratio and MSD have relative larger increasing ratio for a lower heating rate. While the evolution of the relative gyration radius and neck diameter is only sensitive to the temperature.

1. Introduction

The sintering process is a self-propagating synthesis at high-temperature [1], which consolidates the particles into dense polycrystalline materials. As an important traditional technology for material preparation, sintering process had long been extensively studied for its applications in the fields of ceramics, metallurgy, and fire-resistant materials. Several mechanisms, e.g. surface diffusion, grain boundary diffusion, Koch-Friedlander model [2], have been proposed to describe the sintering process. And the surface structure has been regarded as one of the important factors during the sintering. Especially, in the sintering of nanomaterials, the atoms' configuration at the surface plays a more vital role in resultant structure.

Due to the very small dimension at nanoscale, it is difficult to directly observe the sintering process at atomic-level by conventional experimental methods. Therefore, several alternative approaches are proposed to investigate the sintering process of nanocrystalline, among which molecular dynamics (MD) simulation [3–9] has been proved to be a powerful tool. Zhu and Averback [10,11] have investigated the densification process of copper (Cu) nanoparticles based on MD simulations. They have found a new mechanism for grain growth at 1100 K, and the local stress has a significant effect on the sintering process. Grammatikopoulos et al. [12–14] have studied the coalescence of tantalum (Ta) and palladium (Pd) nanoparticles. They have found the wave-liked propagation of the crystallization induced by the rearrangement of atom at atomic scale and proposed an analytical model for nanocluster coalescence. Based on the previous MD simulation results [10,11,15–18], the sintering process at nanoscale is affected by various factors [12–14,19–22], such as temperature, pressure, nanoparticle size, metallic types, lattice orientations and so on.

As heating rate is an important parameter in many thermal treatment processes (e.g. nucleation [23], dissolution [24]), several experiments [25,26] have suggested that heating rate has significant impact on the densification of sintering. Recently, Jiang et al. [27] studied the neck growth in sintering of silver nanoparticles with different heating rates by MD. It was found that the ultrahigh heating rate facilitates the coalescence and melt for hollow particles at a lower temperature level. However, the mechanisms of heating rate effect on sintering are still unclear. In this work, MD simulation is employed to study the aggregation of two Cu nanoparticles with different heating rates. The work is also expected to provide useful insight in metallic coatings [28], nano-polycrystalline materials [14] and nucleation theory [23].

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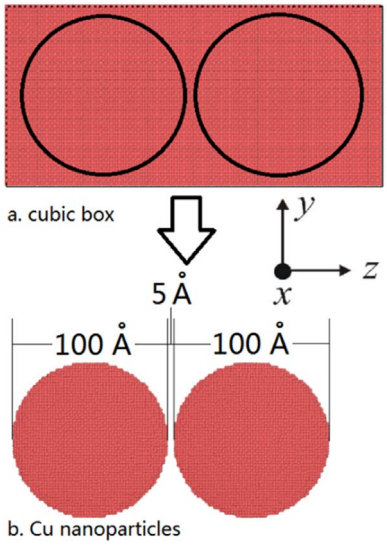

Fig. 1. (a) Illustration of the Cu nanoparticles cut from a cubic single crystal copper. (b) The initial configuration of the simulation system.

2. Model and computational method

2.1. Simulation model

Since this work focuses on heating rate effect on the aggregation of nanoparticles, the classic and common Cu nanoparticles are employed herein. The interaction potential of Cu atoms are described by EAM force field [29]. The initial configuration of the modeling system is consisted of two spherical Cu particles with the same diameter of 100 Å (1 Å=10^{-10} m) that is 89,156 Cu atoms in total, as shown in Fig. 1(b). The distance between the centers of the two spheres is 105 Å. And the spherical particles are cut from a cubic single crystal copper in face center cubic (FCC) 100 orientation with the volume of 252.7×252.7×252.7 $Å^3$, as shown in Fig. 1(a). Periodic boundary conditions are applied in all three directions.

2.2. Computational parameters

MD simulations are performed by using LAMMPS [30] (large-scale atomic/molecular massively parallel simulator). The time-step is set as 1 fs (femto-second) in the simulations. This time-step was tested for energy conservation. Initially, the potential energy of the simulation system is minimized. Then, the simulation system is further equilibrated in the canonical (NVT) ensemble at 300 K for 1,500,000 steps where the temperature is controlled by the Nose-Hoover algorithm. After that, the equilibrated system is linearly heated to 1700 K in 4,000,000, 6,000,000 and 8,000,000 steps, corresponding to the heating rates of 350 K/ns (nanosecond), 233 K/ns and 175 K/ns, respectively. The crystal structure of the nanoparticle is analyzed using an open visualization software OVITO [31].

3. Results and discussions

3.1. Melting temperature

In order to validate the present simulation model and EAM force field, the melting temperature of a single Cu nanoparticle with diameter of 100 Å in the simulation box of 120×120×120 $Å^3$ (44559 Cu atoms) is compared with that of bulk single crystal Cu in a cubic box with 72.2×72.2×72.2 $Å^3$ (32000 Cu atoms), in X×Y×Z dimensions. For both of the two cases, periodic boundary condition is applied in X, Y, Z directions. Both of the structures are linearly heated to 2000 K in 4,000,000 steps after equilibrated at 300 K. The melting temperature of the structure can be deduced based on the transition of the potential energy and specific heat capacity (C_p) during the heating process [32].

As shown in Fig. 2(a), the potential energy of Cu nanoparticle experiences the transition at about 1140 K. This S-shape curve is due to the superheating at a high heating rate. Besides, the maximum specific heat capacity of the Cu nanoparticle with diameter of 100 Å is also located at 1140 K, which further confirms the melting temperature of the Cu nanoparticle is 1140 K. It should be noticed that because of the fluctuation of the potential energy during the heating process, there are couples of peak for the specific heat capacity C_p. While in Fig. 2(b), the transition of the potential energy for cubic single crystal Cu cubic box is insignificant during the heating process, and the maximum C_p locates at 1290 K. A further evidence of the melting is the transformation of the lattice structures in the cubic single crystal Cu, as shown in Fig. 3. Before melting, the lattice structure is perfect FCC. Once the temperature rises to about 1290 K, almost half of the atoms in the cubic Cu change into amorphous structure. Since the melting temperature of bulk Cu is 1358 K [33], closed to the transition temperature of the cubic Cu obtained in this work, it validates the reliability of the present MD simulation.

Due to the periodic boundary condition for cubic single crystal Cu, the structure is similar to the bulk Cu, which results in the melting temperature of cubic Cu closed to that of bulk Cu. The results also suggest that the melting temperature of nanoparticles is lower than that of the bulk material. In general, the formation of nanoparticle is energy consuming that is the mechanical energy or chemical energy transfers to the surface energy, so the nanoparticles usually have higher potential energy than that of their bulk counterpart. Thus, from the energy viewpoint of phase transformation, the melting of nanoparticles needs less energy and they have lower melting temperature.

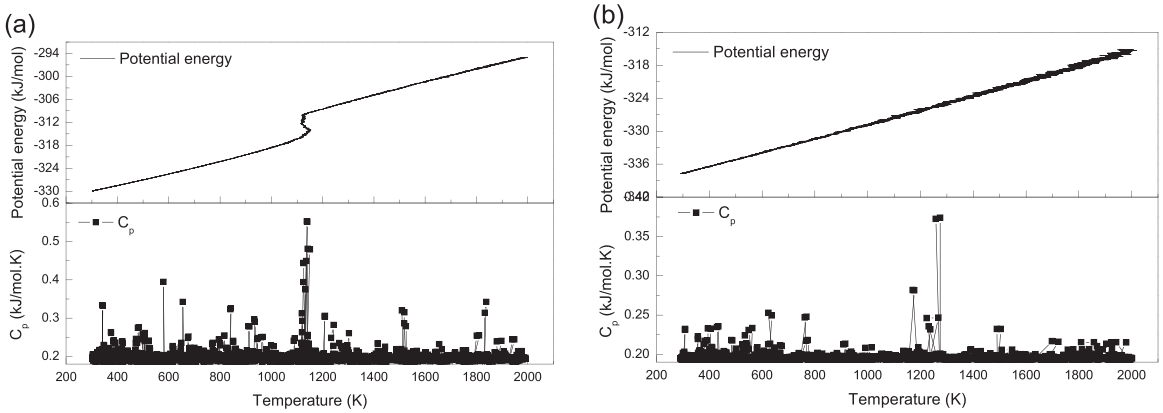

Fig. 2. The evolution of the potential energy and specific heat capacity during the heating process for Cu nanoparticle (a) and bulk single crystal Cu (b).