Vol 10 No 5, May 2001
1009-1963/2001/10(05)/0407-06

Chinese Physics

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and IOP Publishing Ltd

THE INFLUENCE OF GRAIN SIZE AND TEMPERATURE ON THE MECHANICAL DEFORMATION OF NANOCRYSTALLINE MATERIALS: MOLECULAR DYNAMICS SIMULATION

WEN YU-HUA(文玉华), ZHOU FU-XIN(周富信), and LIU YUE-WU(刘曰武)

State Key Laboratory of Nonlinear Mechanics, Institute of Mechanics, Chinese Academy of Sciences, Beijing 100080, *China*

(Received 7 December 2000)

Nanocrystalline (nc) materials are characterized by a typical grain size of 1–100nm. The uniaxial tensile deformation of computer-generated nc samples, with several average grain sizes ranging from 5.38 to 1.79nm, is simulated by using molecular dynamics with the Finnis–Sinclair potential. The influence of grain size and temperature on the mechanical deformation is studied in this paper. The simulated nc samples show a reverse Hall–Petch effect. Grain boundary sliding and motion, as well as grain rotation are mainly responsible for the plastic deformation. At low temperatures, partial dislocation activities play a minor role during the deformation. This role begins to occur at the strain of 5%, and is progressively remarkable with increasing average grain size. However, at elevated temperatures no dislocation activity is detected, and the diffusion of grain boundaries may come into play.

Keywords: nanocrystalline materials, mechanical properties, molecular dynamics
PACC: 3520Y, 6120

I. INTRODUCTION

Nanocrystalline materials are those with grain sizes on the nanometre scale. Typical grain diameters range from 1 to 100 nm, and have two remarkable features, namely, the ultrafine-grain sizes and a large fraction of grain boundary. As the grain size is progressively reduced to the nanometre scale, many mechanical properties, such as hardness, strength and ductility, exhibit a significant difference from those of the coarse-grained counterpart.[1−3] It has been pointed out that size effects may be responsible for specific changes in the mechanisms of deformation.[4] When entering the nanometre regime, the plastic behaviour of metals, which in the bulk situation depends strongly on the dislocation activity, begins to differ substantially because of reduced availability of mobile dislocations in the small nano-grains. Several mechanisms have been proposed to explain the deformation behaviour, based on the presence of porosity, the extensive number of triple junctions, the lack of dislocation activity, grain boundary sliding, and grain rotation, the presence of a softer phase in the boundaries, etc.[5−7] In conventional physical models for crystal plasticity, interfaces represent obstacles for the deformation processes, but in nc materials they are probably responsible for all of the observed plasticity. Computer simulations show that grain boundary sliding is mainly responsible for the deformation of nc materials, but dislocation activity may carry a minor part of the deformation.[8,9] Lu and co-workers have observed experimentally superplastic extensibility of nc copper at room temperature, which originates from a deformation mechanism dominated by grain boundary activities rather than lattice dislocations.[10]

New computational approaches augment the time-consuming search-and-test methods traditionally used for the development of new materials. The atomistic calculations provide a useful tool to analyse structural, mechanical and thermodynamic properties of nc materials, and eventually to design materials of assigned characteristics. The molecular dynamics (MD) approach, as one of the most important methods of atomistic simulations, provides the phase-space trajectories of particles through the solution of Newton's equation, thereby shedding light on how atomic level processes can lead to macroscopic phenomena, and so it can help in understanding the relation between structure and overall properties. MD simulation could display trajectories of atomic motion, and may be used to carry out a variety of observations similar to those in experiments. Especially, those related with atoms can be easily acquired through the MD simulation but cannot in experiment. MD simulation, not limited by the synthesis, thermal history and testing technique

of the samples, may be used to elucidate the experimental results in theory, and make up for the shortage of the experiments. In MD simulation of nc materials, not only the correct choice of the potential, but also the construction of the atomic model are essential.

In this paper, we present MD simulations of the tensile deformation behaviour of nc materials. We investigate the structures of the grain boundary and the interior of grains during the deformation, and study the influence of average grain sizes and temperature on the mechanical properties.

II. SIMULATION METHODS

A. Generation of the nc 'samples'

To computer simulate the deformation of nc materials, we construct nc 'samples' with structures reasonably similar to those observed experimentally: essentially the equiaxed dislocation-free grains separated by narrow grain boundaries. Four samples, each containing approximately 55 300 atoms with 8, 27, 64 and 216 grains (Cu:5.38, Cu:3.59, Cu:2.69, and Cu:1.79), which represent average grain sizes ranging from 5.38 to 1.79nm, are constructed by filling the simulation cell volume of an 8.67 nm cube. The grains are produced by a Voronoi polyhedron construction.[11] A set of grain centres are chosen randomly, and the part of space closer to a given centre than to any other centres is filled with atoms in a randomly rotated fcc lattice. In order to eliminate unfavourable configurations in grain boundaries, the samples are relaxed by running a 50 ps MD simulation at 300 K and 0 Pa, with a Finnis–Sinclair potential[12,13] in the Parrinello–Rahman approach[14] under periodic boundary conditions, which replicate the cube infinitely in all three spatial directions. The atomic trajectories are followed by integrating the Newton's equation of motion for each atom with the Frog-Leap algorithm.[15] The time-step in simulation is $\tau_0 = 5 \times 10^{-15}$s, the energy unit 1.0 eV, and the length unit $a_0 = 3.615 \times 10^{-10}$ m.

B. Deformations of samples

The main part of the simulation is the slow uniaxial tensile deformation while minimizing the energy with respect to all atomic coordinates under different conditions. The systems can be deformed by rescaling the simulation cell and the coordinates along the X direction in space. At the same time, the box sizes in the Y and Z directions are allowed to shrink to keep the transversal component of the stress (σ_{yy} and σ_{zz}) close to zero. In other words, the samples are slowly loaded along the X axis until a total strain of 12% is reached, and the normal stresses along the Y and Z axes are simultaneously held on 0 Pa. All simulations of the mechanical deformation may be divided into two parts. Firstly, we simulate the deformation of nc samples with four different average grain sizes, i.e. 5.38, 3.59, 2.69 and 1.79nm at 300 K. Secondly, we simulate the deformation of the Cu:5.38 sample at 300, 600, 900 and 1200 K.

C. Analysis of the structures

To facilitate the structural analysis of simulations, we identify which atoms are located at grain boundaries and which are inside the grains, by determining the local crystalline order using the common neighbour analysis.[16] We may define three categories of atoms: 'fcc', atoms having a local fcc order are considered to be inside the grains; 'hcp', atoms having a local hcp order are classified as stacking faults; and all other atoms ('other' atoms) are considered as belonging to grain boundaries. Radial distribution functions[17] are a useful way to describe the structure of a system, and are used in this paper.

III. RESULTS AND DISCUSSION

A. Structure and energy distribution

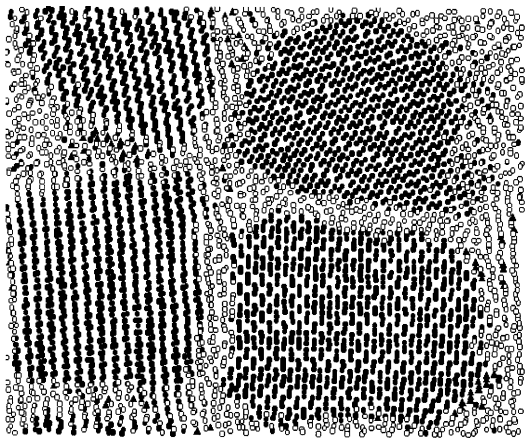


Fig.1. Snapshots of the simulated nc Cu:5.38 sample before deformation, which contains eight grains and about 55 300 atoms, giving the mean grain diameter of 5.38 nm. Solid circles, triangles and open circles denote 'fcc' atoms, 'hcp' atoms and 'other' atoms, respectively.

The atomic structure of the Cu:5.38 sample, which is relaxed by running a 50 ps molecular dynamics simulation at 300 K and 0 Pa before the deformation, is illustrated in Fig.1 in cross section through

four of the eight grains. It is clearly seen that all 'fcc' atoms are located in the interior of the grains, and the 'other' atoms are uniformly distributed in the fringe of the grains. All 'hcp' atoms are dispersed in the grain boundaries and there is no 'hcp' atom in the grain interior. This shows that the structural analysis is appropriate, and the computer-generated samples are consistent with those observed experimentally.

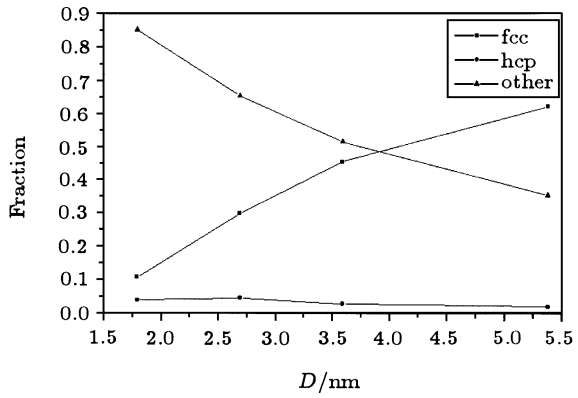


Fig.2. The structural variation of nc Cu as a function of grain size.



Fig.3. The contours of equal potential energy per atom corresponding to the cross section in Fig.1.

Figure 2 displays the structural analysis results of four samples. From this figure, we see a decrease of the fraction of the 'fcc' atoms (from 54% to 12.5%) and a corresponding increase of that of the 'other' atoms (from 33.5% to 82.8%) as the average grain size is reduced. But that of the 'hcp' atoms remains approximately the same (between 2.5% and 5.5%). This indicates that the volume fraction of grain boundaries increases significantly with the decrease of average grain size. Figure 3 shows the contours of equal potential energy per atom corresponding to the cross section in Fig.1. As we would expect, the areas of high energy coincide with the grain boundaries, and the potential energy of the interior of grains is lower than that of the grain boundaries, but slightly higher than that of the perfect crystal.

B. The grain size effect

The grain size has an important influence on the mechanical properties of polycrystalline materials. As the grain size is reduced, the strength and hardness of the materials evidently increase, but in the nanometre regime, we have not understood how they are changed. As shown in Fig.2, the fraction of grain boundaries increases significantly with the decrease of the average grain size below 6 nm. According to the dislocation theory, it is reasonable to conclude that, for a grain size smaller than 10 nm, the nucleation and motion of dislocations will be severely restricted and the grain boundaries may play a crucial role in the deformation.

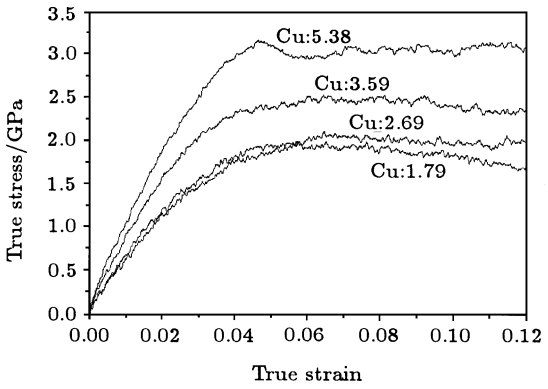


Fig.4. The true stress versus the true strain for four samples.

Figure 4 shows the simulated true stress–strain curves of nc samples at 300 K. A significant strength increase is clearly observed as the average grain size increases from 1.79 to 5.38nm. Compared with the experimental results of nc Cu, the stress of the simulated Cu is distinctly higher, which may come from a variety of effects, such as pores (and larger flaws), lattice or surface defects and impurities of nc materials in experiment. These depend on the way in which these materials are synthesized and processed and, as a result, these factors reduce the strength of the materials. The flow stress of nc samples, which is defined as the maximal stress of the stress–strain curves, has increased as the average grain size increases. The result shows a reverse Hall–Petch relationship, which is not consistent with that of the coarse-grained counterpart materials whose mechanical response is dominated by

a high pre-existing dislocation structure.

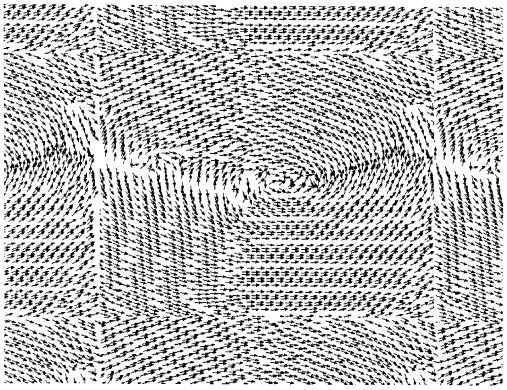


Fig.5. The displacement vector plot of the simulated nc Cu:5.38 from the undeformed to 1% strain, revealing the presence of grain rotation, grain boundary sliding and grain boundary motion during the deformation.

To explore the reasons resulting in a reverse Hall–Petch relationship, we plot the displacement vector graph of a deformed Cu:5.38 sample, as shown in Fig.5, corresponding to the cross section in Fig.1. One may see the presence of grain rotation, grain boundary sliding and motion during the plastic deformations, which have been observed in other computer simulations.[8,9,18] We have analysed the structure of the Cu:5.38 sample during the deformation and show the cross section plot of this for a strain of 12%, as shown in Fig.6. We clearly see some stacking faults, which do not extend through the crystalline plane, left behind by partial dislocation activities that have run through the grains during the deformation processes. Figure 7 shows the fraction of the 'hcp' atoms as a function of true strain. When the strain increases from 0% to 12%, the enhancement in 'hcp' atomic percentage (7.94% in Cu:5.38, 4.35% in Cu:3.59, 3.31% in Cu:2.69 and 2.15% in Cu:1.79) has dropped with the decrease of average grain size. With the strain smaller than 4%, there is an insignificant change in the number of the 'hcp' atoms in nc samples; this means that almost all of the deformation has been carried out through grain boundary sliding, grain rotation and grain boundary motion. The stacking faults begin to appear at about 5% deformation when some partial dislocation activities have carried a minor part of deformation. The percentage of stacking faults have decreased as the grain size is reduced, because a larger fraction of atoms belongs to grain boundaries, and the grain boundary sliding and grain rotation become easier to take place.

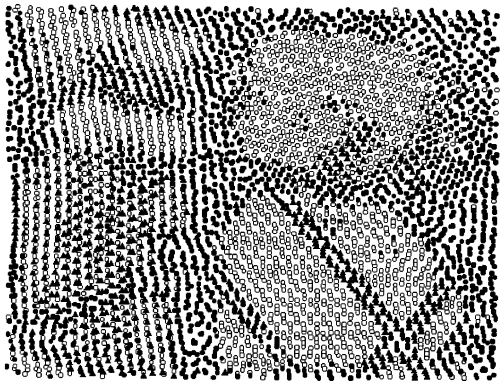


Fig.6. Snapshots of simulated Cu:5.38 sample with 12% strain. Open circles, triangles and solid circles represent 'fcc' atoms, 'hcp' atoms, and 'other' atoms, respectively. We may clearly see some stacking faults in the grains.

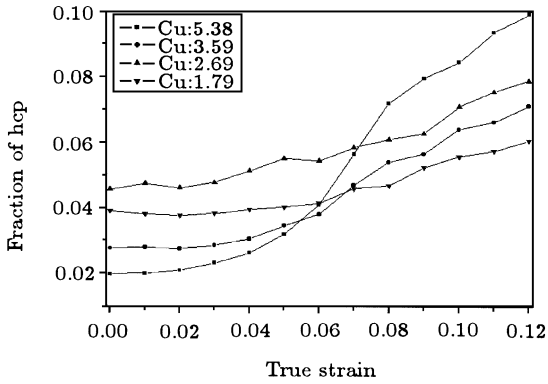


Fig.7. The strain dependence of 'hcp' atom percentage for different grain sizes.

C. The temperature effect

Temperature has a significant influence on the microstructure and the mechanical properties of materials. At low temperatures, the boundaries act as obstacles to the easy movement of dislocations, and thereby they provide strengthening through the Hall–Petch relation. However, the grain boundaries represent a source of weakness at high temperatures, both because of their ability to promote premature failure through the development of intercrystalline cracks and cavities, and because they may contribute directly to the overall deformation through the occurrence of grain boundary sliding. In nanoscopic range, the grain size is sufficiently small, of the order of only a few nanometres, so that the behaviour of boundaries may account for essentially all of the deformation under all testing conditions.

In order to study the temperature influence on mechanical properties, we simulate the deformation behaviour at different temperatures. Figure 8 shows

the radial distribution functions (RDFs) of the undeformed Cu:5.38 sample at 300, 600, 900 and 1200 K. One may see that the degree of disorder has distinctly increased with the increasing temperature of the system. It is worth noticing that at 1200 K, the second peak of the RDF has already been broadened and presents a low intensity, indicating that the structure of the Cu:5.38 sample is highly disordered but not completely amorphous, meaning that it is not liquid-like or amorphous. Figure 9 displays the simulated true stress–strain curve at different temperatures. We clearly see a decrease of flow stress at elevated temperatures, which may be due to the fact that at high temperatures the average interatomic spacings of the nc system are increased relative to those at low temperatures if the atomic potential is assumed to be the same at any temperature. Further structural analyses indicate that at high temperatures (>900 K) there are few dislocation activities in the grains, and atoms in grain boundaries with high energy make the sliding and motion of grain boundary and the grain rotation easier to take place. Another deformation mechanism may be grain boundary diffusion, in which the diffusivity of nc materials is much higher than that of coarse-grained counterpart at high temperatures. This mechanism has already been observed experimentally to be effective in nanophase ceramics,[19] and the related simulations are still to be carried out.

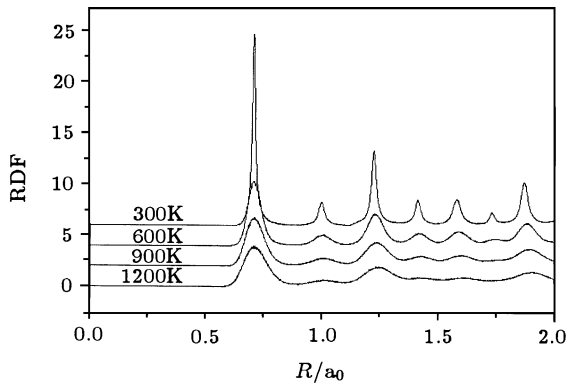


Fig.8. The radial distribution functions of Cu:5.38 sample at different temperatures.

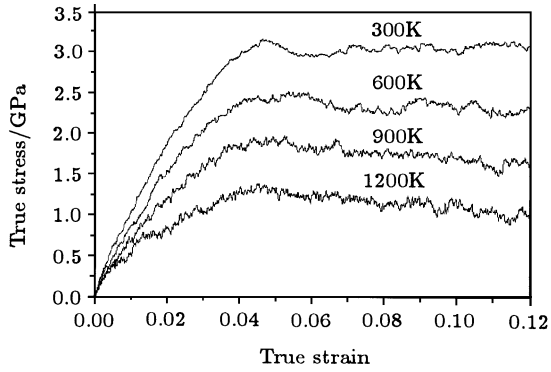


Fig.9. The true stress versus the true strain for Cu:5.38 samples at different temperatures.

IV. CONCLUSIONS

We have performed molecular dynamics simulations of the behaviour of the deformation of nc copper samples with different grain sizes, and studied the influence of grain size and temperature on the mechanical properties. We have analysed the structure of nc samples during the deformation using common neighbour analysis. From the above simulated results, the following conclusions can be made:

1. The strength, as well as the flow stress, has distinctly increased as the grain size increases, indicating a reverse Hall–Petch effect. During the plastic deformation, grain boundary sliding and motion, as well as grain rotation are identified. They are mainly responsible for the plastic deformation, and a minor part is carried out by partial dislocation activities in the grains. The dislocation activities begin to play a role at the strain of 5%, and this role is progressively distinct with increasing grain size.

2. Temperature has a significance influence on the mechanical deformation. With the increase of temperature, the flow stress decreases distinctly, and the nc material shows a trend of softening. The main deformation mechanisms are the sliding and motion of grain boundary and grain rotation. At low temperatures, partial dislocation activities play a minor role. However, at elevated temperatures there is essentially no generation of stacking faults, indicating that no dislocation activities occur in the grains, and the diffusion of grain boundary may play a certain role.

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