Molecular Dynamics Simulation of the Grain Growth in Nano-Grained Metallic Polycrystals

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The grain growth phenomenon in a polycrystalline metal was studied by the molecular dynamics method. The time evolutions of the grain boundary network in the nano-grained polycrystals composed of 64 grains were analyzed by incorporating the boundary curvature and the grain boundary energy composed by the interatomic potentials. The grain growth phenomenon accompanied by the T1 and T2 processes was successfully observed. The rate of the grain growth was fairly close to the parabolic law. The grain boundaries were dominantly composed by random boundaries, but a number of coincidence boundaries were also observed.

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1. Introduction

Microstructures of polycrystalline metals and ceramics are fundamental characteristics to control and improve the material properties. Prediction of the grain boundary structure after the manufacturing processes is important for materials design and processing.¹⁾ The grain boundaries in a polycrystalline solid constitute a complex network structure which shows a time evolution toward minimizing the grain boundary energy at elevated temperatures. As the results, small grains disappear and the mean size of grains increases with time. Such grain growth process is influenced by not only the boundary topology but also the boundary misorientation and microstructures including point defects and dislocations.

In order to express the grain growth phenomenon by using simple mathematical formulae, various studies have been developed for many years. The mean field approaches were made by Hillert²⁾ and Louat³⁾ during 1960s and 1970s. Srolovitz *et al.*⁴⁾ used the Monte Carlo method (so called Potts model) in 1980s, and Anderson *et al.*⁵⁾ extended the method to three-dimensional (3D) problems. Atkinson⁶⁾ reviewed such classical theories, models and computer simulations on the grain growth phenomena. On the other hand, the vertex model⁷⁾ has also been applied to the coarse-grained problems including grain growth, liquid foams, and biological textures. Recently, Wakai *et al.*⁸⁾ extended the vertex model to the 3D curved grain boundaries by using the surface evolver method.⁹⁾ Most of these methods, however, assume the grain as a homogeneous media or even a vacant space, and the grain boundary as a simple mathematical surface without any microstructures. It is easily understood that such assumptions are not always acceptable in the atomistic level.

Molecular dynamics (MD) simulation appears to be useful for analyzing the grain boundary kinetics by incorporating the microstructures in atomistic levels. We can follow all atomic motions at the grain boundary during the simulation. Recently, Upumanyu¹⁰⁾ made an MD simulation on the grain boundary mobility by using the half-loop grain boundary model and the Lennard-Jones potential. However, due to

the limitation of the computer ability, simulation of polycrystalline matter has been carried out only recently,¹¹⁻¹⁴⁾ and not applied to the grain growth problems yet. In this paper, the present authors simulated the grain growth phenomenon in a nano-grained polycrystalline metal. This method takes the atomistic microstructures including the crystal lattice, boundary misorientation, curved interfaces, and triple junctions into account which are frequently ignored in other methods.

2. Molecular Dynamics

The initial structure of the present model is composed of 64 grains in the nanometer scale. All grains are two-dimensionally arranged in the rectangular 3D basic cell. The size of the basic cell is $25 \times 25 \times 1.4 \text{ nm}^3$, and the periodic boundary conditions were applied. Partitioning of the space into the grains was carried out by the Voronoi construction.¹³⁾ Initially, all grains have almost the same size of about 3.5 nm in diameter and four- to nine-sided shapes in the x - y plane. The crystallographic orientation of each grain has common rotation axis of $\{311\}$ toward z axis and randomly selected rotation angle in the x - y plane. Such limitation of the crystal orientation is necessary to satisfy the boundary condition toward the z axis.

The target material of the present study is iron. The many-body interatomic potential of Finnis and Sinclair¹⁵⁾ was used. The MD simulation was started at the room temperature and heated up to 1680 K, just above the $\gamma - \delta$ transition temperature, at the atmospheric pressure. After a short relaxation time of several picoseconds, constant NPT simulation using the Nosé thermostat was carried out for more than 500 ps. The resulted crystal structure in the simulated sample was b.c.c. We used the parallelized MD code¹⁶⁾ and the workstation cluster HIT HPC-HP2SC/Myrinet composed of 12 CPUs. Three independent simulations were carried out for slightly different initial structures.

3. Results

3.1 Global features of the grain growth

The grain growth phenomenon has started during the heating and relaxation processes of the computational sample, hence several grains in the initial structure were already lost before starting the constant temperature simulation. For convenience, we define the origin of the simulation time t as the time of the constant NPT simulation started. In Fig. 1 we show the snapshots of the simulated samples at different stages in the same simulation. In the figure, only the atoms having higher potential energy were drawn in order to clarify the grain boundary networks. At the early stage of the simulation (a), several grains were growing by corroding the

Fig. 1 Snapshots of the simulated sample projected to the $x - y$ plane at $t = 20$ ps (a) and 150 ps (b). Only the atoms having the higher potential energy are displayed.

neighbors, and hence the grain size distribution was getting broader than the initial structure. At $t = 150$ ps (b), the number of grains reduced to less than 20 and the mean grain size increased. The number of grains decreases to less than 10 at about $t \approx 500$ ps in the present simulation.

It is well known that the rate of the grain growth can be expressed by the parabolic law, $r^2 \propto t$, where r is the mean radius of the grains. In Fig. 2 we plot the mean area of the simulated grains in the $x - y$ plane as a function of time for three independent simulations. They show almost the same time dependence close to the parabolic law. This result is quite interesting because it means that the empirical law found in the macroscopic grain growth phenomena is also applicable in the nanometer scale.

3.2 Topological variation of the grain boundary network

The network structure of the grain boundary changes its topological form during the grain growth phenomenon. In the case of the 2D grain growth, topological changes of the network are expressed by two elemental processes: the grain switching (T1) and the disappearance of a grain (T2).⁶⁾ A number of T2 processes were observed in the present simulation whenever grains disappeared. The T1 processes, on the other hand, were not frequently observed compared with the T2 process. One example of the simulated T1 process is shown in Fig. 3. In this case, two grains of pentagonal shape (we call them as A and B) were initially contacting to each other (a), and then separated after the T1 process (b). As the result, grain boundary between A and B vanished and a new grain boundary between the other two grains (we call them as C and D) was formed. The number of the sides of concerning grains varied due to this process: A and B lost one and C and D gained one, respectively.

It should be noted that the curvature of the grain boundary does not always obey to the macroscopic picture that convex for a grain having less than 6 sides and concave for greater than 6. Such deviation of the boundary curvature in the present model may be caused by the fact that the angles at the triple junction are not always 120° due to the difference in the grain boundary energies.

Fig. 2 Time variation of the mean grain area in the $x - y$ plane for three different simulations.

Fig. 3 Snapshots of the simulated sample before (a) and after (b) the T1 process. Atoms having the higher potential energy are painted in dark gray to clarify the grain boundary positions.

3.3 Grain boundary characteristics

In the present model, crystal orientation of each grain was selected randomly within the limitation of the periodic boundary condition, and most of the grain boundaries are so called random boundaries. However, we observed a few boundaries having smaller thickness than the others as shown in Fig. 1. They are the low energy boundaries. By analyzing the misorientation angles of such boundaries, we found some of them are the $\Sigma 3$ boundaries (rotational angle of 146.4° around $\langle 311 \rangle$ axis). Although the random boundary is dominant in all cases, coincidence boundaries were also observed by changing their positions during the grain growth. Coincidence boundaries such as $\Sigma 3$ are constructed when contacting the grains in the special angular relations, and seem to remain longer than the random boundaries.

4. Discussion

As shown in above, the present MD simulation seems to reproduce the grain growth phenomenon successfully. However, we should note that the present simulation covers very

narrow scales in space and time less than 25 nm and about 1 ns, respectively. The reliability of the present simulation must be checked by comparing with experimental results. As one of such comparisons, the authors analyze the Aboav-Weaire law which was originally observed in metallic samples experimentally.^{17,18)} According to this law, the product of the number of sides n of a grain and the mean number of sides of surrounding grains, $m(n)$, is proportional to n . The results of the MD simulation are plotted in Fig. 4 for different stages of the grain growth. Although the small deviations due to the difficulty of analysis and limited numbers of grains are recognized, a linear relation is successfully reproduced. This can be an evidence for the similarity between the simulated and real grain growth phenomena.

Another discussion must be made on the grain size dependence. Kim *et al.*¹⁹⁾ estimated the volume fractions of the grain, grain boundary, triple line, and the quadruple junction in 3D polycrystals as a function of the grain size, and showed the grain boundary region is dominant in nano-scale polycrystals. Although the present model has a two-dimensional grain arrangement and differs to the Kim's model, we should note

Fig. 4 The Aboav-Weaire relation in the simulated sample.

that the effect of the grain boundary is possibly emphasized than that in larger scales.

On the other hand, the present method has advantages over the traditional methods. All information on the atomic configurations at the grain boundary and triple junctions is available. We can also analyze misorientation and curvature of all grain boundaries. As shown in above, coincidence boundaries were observed in the present simulation similar to the experiments. This is quite important for the grain growth study because the boundary energy and the boundary curvature are the fundamental driving forces of the phenomenon.

5. Summary

The present MD simulation was successful for reproducing the grain growth phenomenon in metallic polycrystals in the atomistic scale. Our method incorporates fundamental driving forces of the grain growth: grain boundary energy and

the boundary curvature. The simulated results are close to the well known characteristics of the grain growth including the parabolic low and T1 and T2 processes. MD simulation is applicable to not only the grain growth phenomenon but also the formation of the materials microstructures. In conclusion, the present authors stress that the MD simulation is useful for investigating the atomistic process of polycrystalline materials especially at high temperatures. It would be interesting to extend the present model to various conditions and the usefulness and validity will be tested in a wider base.

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