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Computer simulation of normal grain growth in polycrystalline thin films

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A modified Monte Carlo method is proposed to simulate the two-dimensional normal grain growth in polycrystalline thin films. With the newly modified method, not only the simulation efficiency is improved but also the simulated time exponent of grain growth attained $n = 0.49 \pm 0.01$, which is very close to the theoretical value for steady grain growth $n = 0.5$. Simulation of the complete process of normal grain growth including the steady state is made possible by means of the present method. The grain size distribution in the simulated thin films was found to vary continuously and slowly with time, the gamma and the Hillert functions may be two of the expression forms during its transition, and the latter corresponds to quasi steady grain growth. The so called "self-similarity" of the grain size distribution during the normal grain growth in two-dimensions is also discussed according to the simulation results.

1. Introduction

The process of normal grain growth in polycrystalline thin films seems to attract less attention than that in three-dimensional materials, but it plays an important role in the theoretical research of microstructure evolution and in the prediction as well as control in practice for low-dimensional materials. In the kinetics of normal grain growth, the Grain Size Distribution (GSD) can reflect uniquely the distribution state of the grain size in the whole system at a certain time. It displays the instantaneous evolution of grain size configuration more precisely than the average grain size. Some experimental results [1–4] showed that the shape of GSD keeps basically unchanged, or so called self-similarity [5], during the process of normal grain growth in polycrystalline thin films, but it has been some time since attempts were made to deduce the Grain Size Distribution Function (GSDF) from theory [5–7]. One of the main conclusions of the famous "mean field theory" of Hillert [6] is the GSDF in two-dimensions for the thin films which has the form as follows:

$$F(u) = (2e)^2 \frac{2u}{(2-u)^4} \exp\left(-\frac{4}{2-u}\right) \qquad (1)$$

where u is the relative grain size. Unfortunately, none of the experimental data have been found up to now to be consistent with the Hillert distribution. Besides the theoretical predictions, the lognormal or the gamma function are often applied to fit the experimental GSD of normal grain growth [5]. It is known that any practical condition of grain growth in polycrystalline materials deviate to a varying degree from the assumptions of theoretical models, so experimental results alone will not prove conclusive. Computer simulation is the way to solve the difficulty that it is impossible for the practical process to attain the ideal conditions of the models. Determination of an appropriate simulation method should be the first step to correctly describe the complete process of normal grain growth in thin films.

Since 1980s the Monte Carlo (MC) method has been applied by more and more researchers to simulate grain growth in polycrystalline materials [8–10]. It is a direct and effective way to describe the kinetics and topology of grain growth. However, there are some limitations existing in the algorithm which is being used presently, influencing the accuracy and efficiency of the grain growth simulation, especially in the field of the grain growth kinetics [11]. In the present paper a modified MC method will be built in order to investigate the characteristics of normal grain growth in the polycrystalline thin films.

2. Simulation procedure

In the MC method for simulation of two-dimensional grain growth, the plane is divided into a large number of tiny-sized hexagons or squares which act as the basic units of the grain structure and are called as "the sites". The sites with the same orientation construct a

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grain, the grain boundary lies between the sites with different orientations. Reorientation of the site close to the grain boundary results in the motion of the grain boundary, i.e., grain growth. As far as the micro physical base of the grain boundary motion is concerned, the grain boundary mobility only results from the process that the atoms close to the grain boundary jump into the nearest grain interior, so the equivalent mobility rate of the grain boundary in the MC simulation is proportional to the efficient reorientation probability of the sites. In the original method [8,9], the reorientation of a site is one of the possible orientation states of the whole system, the equivalent mobility rate of the grain boundary decreases with the decrease of the efficient reorientation probability of the sites, which does not agree with the physical base concerning the motion of the grain boundary, and as a result, influences inevitably the simulated grain growth exponent and meanwhile consumes too much calculation time. Besides, each cycling step in the original method [8–10], named as a MCS, contains the re-orientating attempts of all sites of the system, but the site to re-orientate is selected randomly; this treatment has no reasonable meaning but occupies the CPU time in vain.

Accordingly, three points are taken into consideration by present authors to build a MC method for simulation of normal grain growth in thin films, as described below:

(1) The reorientation of a site is determined as the same as one of its nearest neighbouring sites; this procedure is quite different from the original method which selected one of the total orientation states in the whole system [8, 9], and the physical base of grain growth is obeyed. What is more, the phenomena of the "grain coarsening" and "re-nucleating" which may occur in the original method, will be avoided.

(2) The principle of "site selecting and re-orientating one by one" is used in each MCS, the simulation efficiency is thus improved compared to the original method [8, 9].

(3) If the energy connected with the orientation configuration of sites reduces after the site reorientating, the site re-orientates successfully; if the energy increases, the site keeps its original state; if there is no energy change after the site re-orientating, the site changes the orientation state or keeps the original state with the same probability. This criterion of micro migration of grain boundary is more accurate than the original method [8, 9].

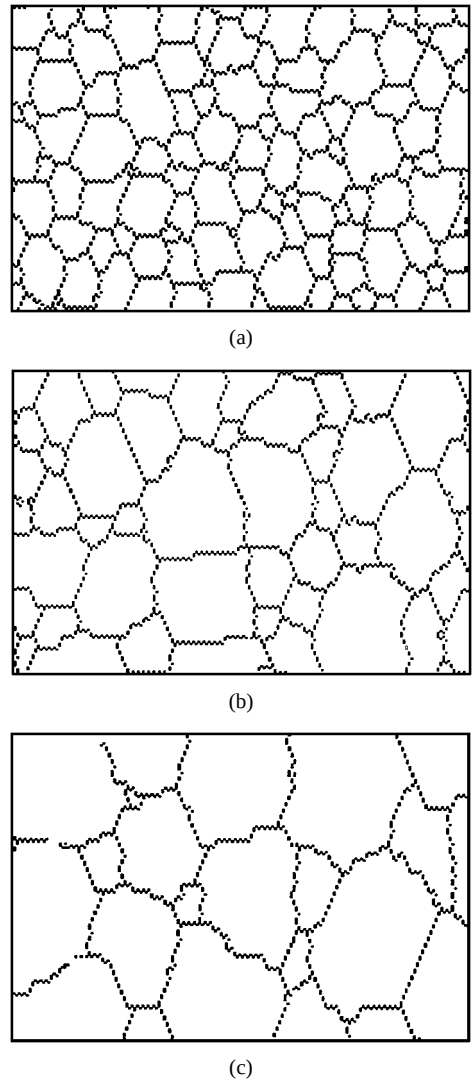

Figure 1 The instantaneous evolution of the simulated grain structure in polycrystalline thin films: (a) 100 MCS, (b) 500 MCS and (c) 800 MCS.

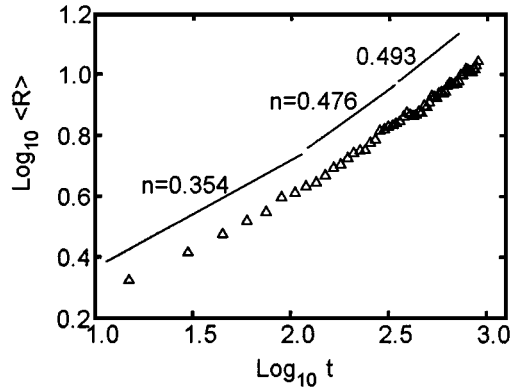

Figure 2 Changes of the average grain size with time. The time exponents marked in the figure are mean values in different grain growth periods.

3. Simulation results

3.1. The time exponent of normal grain growth

Fig. 1 displays the instantaneous evolution of the simulated grain structure in polycrystalline thin films. It was observed that normal grain growth is always kept during the long time simulation and the "grain coarsening" does not occur. Fig. 2 shows changes of the average grain size measured from area with time, the time exponent was observed to increase slowly with time, the exponents marked in the figure were mean values in different periods. In the later period of grain growth after 600 MCS, the time exponent obtained from the present method attained $n = 0.493$ (being very close to the theoretical value $n = 0.5$ [5–7]) and kept almost unchanged later on, indicating that the simulated grain growth attained the steady state. Therefore, the present MC method is suitable to simulate more completely

normal grain growth in thin films than other ones before [8–10], which obtained grain growth exponents more deviated from the theoretical value.

3.2. Grain size distribution

The GSD at time $t = 30$ MCS after which the configuration of compact grains has formed, is shown in the histogram of Fig. 3a, comparing with the lognormal, gamma and Hillert functions. It can be seen clearly that the gamma function agreed much better with the GSD than the other theoretical functions. Further, during the simulated range of $t < 400$ MCS, the GSDs can be expressed properly by the gamma function as indicated in Fig. 3b. As the grain growth proceeded, the GSD deviated gradually from the gamma function, which became more obvious when the simulating time was longer than

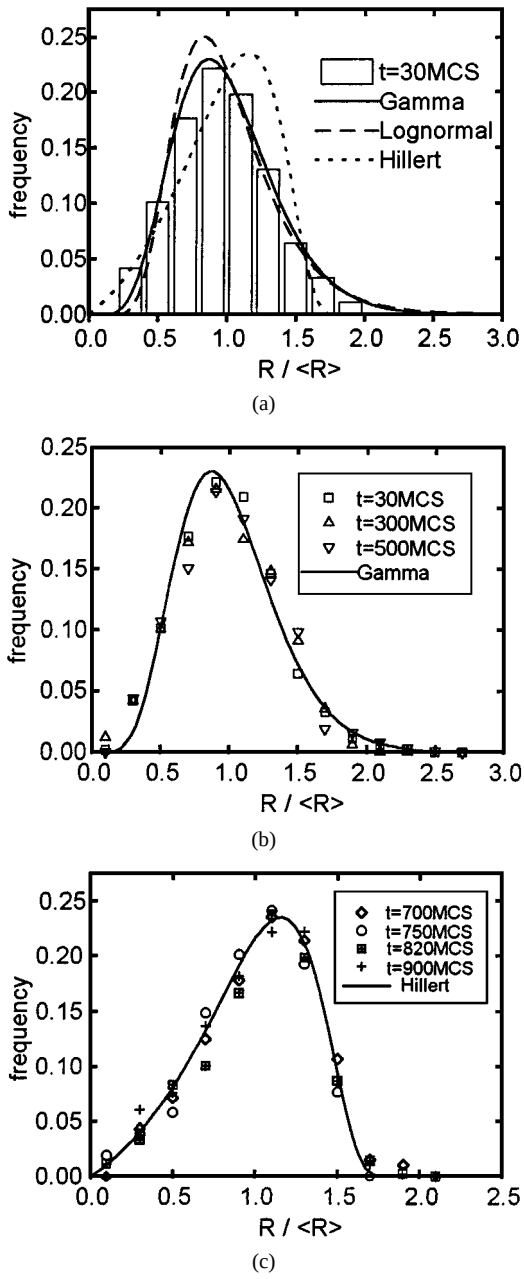

Figure 3 (a) The GSD at $t = 30$ MCS, comparing with the lognormal, the gamma and the Hillert distribution function [6], (b) The GSDs in the range of $t < 400$ MCS and (c) The GSDs in the range of $600 < t < 950$ MCS, which coincide with the Hillert function [6].

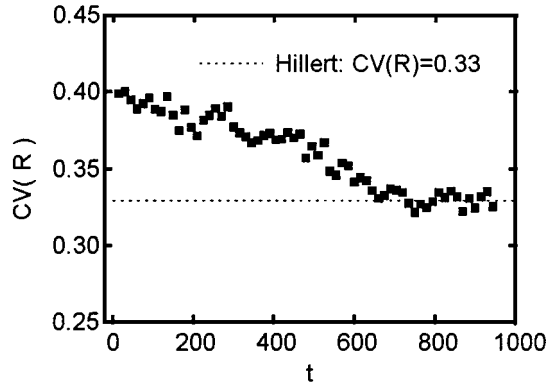

Figure 4 Changes of the variation coefficient CV(R) with the simulating time during the long time simulation.

550 MCS, from which moment the GSD exhibited the right-screwed characteristic. When the simulating time $t > 600$ MCS, the GSDs seemed to transit to the Hillert function as shown in Fig. 3c, then the GSDs changed little until the end of the simulation of grain growth at the time when the number of grains decreased to fewer than 150. The above simulation results showed that the GSD does not keep an unique shape if the simulated process of normal grain growth is sufficiently complete, in other words, the "self-similarity" of the GSD during the complete normal grain growth in thin films does not really exist, the GSD varies continuously and slowly with time. To make this clearer, the variation coefficient CV(R) of the GSD was used to reveal precisely the evolution of the distribution. Fig. 4 shows the changes of CV(R) with the simulating time during the long period of normal grain growth. The variation coefficient varied from 0.42 to 0.32 within the regime examined, but in a small interval the value undulated weakly, which may be the reason for the relative stability of the GSD in this short period. After the simulating time exceeded 600 MCS, CV(R) decreased from the higher value to a value about 0.33 and basically stabilized at this value until the end of the simulation. As is known, the value $CV(R) = 0.33$ is just the variation coefficient of the Hillert distribution [6].

4. Discussion

It is considered by many people that the invariability of the GSD is one of the characteristics of normal grain growth [1–7] since little change in GSD has been observed in some experiments [1–4] and obtained from some computer simulations [8–10]. However, it should be noted that these results are restricted by the experimental conditions and by some simulation algorithms, thus reflect only the features of certain stages of normal grain growth. As a result, the so called time-invariant GSD both from the experiments and the limited simulations, cannot represent the GSD of the more complete normal grain growth. As far as the GSD of the whole process of normal grain growth is concerned, it is not rigorous to define it as "self-similar" [5], because the GSD varies continuously and slowly with time, neither the gamma function nor the Hillert function can describe the variable distribution appropriately. Since

the GSD with the shape similar to the gamma function usually remain for a long time with little change in the variation coefficient, the gamma function is often taken as the "self-similar" distribution of normal grain growth both in experiments and some simulations [5], but it is not the distribution form of the steady grain growth. The GSD of the quasi steady state with the growth exponent very close to the theoretical value $n = 0.5$, has the form approximately consistent with the Hillert function (see Fig. 3c). This means that the Hillert distribution probably exists and is one of the distribution forms of the complete grain growth, corresponding to the quasi steady state. The reason that the Hillert distribution has never been reported to obtain is considered by the authors to lie in two aspects: one is that it is very difficult in fact to keep the thin-film polycrystalline material performing normal grain growth for sufficiently long; the other is that some two-dimensional computer simulations restrain the grain growth to achieve the steady state, so it is difficult for the simulated growth exponent to attain the theoretical value $n = 0.5$.

The presently modified MC method allows the study of the relatively complete process of normal grain growth in two-dimensions, and it is found that the gamma and the Hillert functions are two similar forms of the GSD during its transition. It is questioned by the authors whether there exists a reasonable function containing more than two variables that can describe precisely the slow evolution of GSD in the complete process of normal grain growth in thin films. Such theoretical study should be further developed.

5. Conclusions

The following conclusions were obtained from the present paper:

(1) The modified MC method was proposed to simulate the normal grain growth in polycrystalline thin films;

(2) The simulated time exponent of normal grain growth increased with time and nearly attained the theoretical value $n = 0.5$, indicating the possibility to study the complete process of grain growth in two-dimensions;

(3) The grain size distribution of the complete grain growth in thin films was found to vary continuously and slowly with the simulating time, the gamma and the Hillert functions may be two forms during its transition, and the latter corresponded to the quasi steady grain growth.

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