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Discussion on the Hillert Theory of Normal Grain Growth with a Modified Monte Carlo Simulation

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Abstract: On the basis of analyzing some limitations in the existing algorithm, a modified Monte Carlo method was proposed to simulate two-dimensional normal grain growth. With the modified method, the simulated time exponent of grain growth attained n=0.49 ± 0.01, which is very close to the theoretical value of the steady grain growth n=0.5, indicating the possibility to investigate the total process of normal grain growth. The relation between the Hillert and the von Neumann equations were studied and identified, the Hillert's basic equation has been found to hold during the normal grain growth. The grain size distribution was found to vary continuously and slowly with the simulated time in the total growth process, the lognormal and the Hillert functions may be two types of the expression forms during its transition, and the later seemingly corresponds at the distribution of the steady stage were $n \approx 0.50$.

Key words: Monte Carlo simulation, Hillert theory of normal grain growth, von Neumann equation, grain size distribution

Research on the grain growth is important and meaningful not only in the field of metallurgy but also in the geology (e.g. the petrology) and biology (e.g. the histology)[1]. According to the understanding of the grain growth process, it is possible to control and improve the properties of materials. So, it has been a long time since people devoted into investigating the grain growth theoretically and experimently. Many theoretical models of grain growth have been proposed till now. The Hillert theory of normal grain growth[2] is one of the most widely accepted classical models among them, which will be explained in section 2.

Any practical process of grain growth in polycrystalline materials deviates to a varying degree from the assumptions of the theoretical models, so it is probable to obtain indefinite conclusions from the experimental identification alone. Computer simulation is just the way to solve the difficulty that it is impossible for the practical process to attain the ideal conditions of the models, hence more and more reseachers[3 ~ 8] adopt this technique to study the grain growth under the theoretical conditions. Monte Carlo(abbreviated as MC hereafter) simulation is a direct way to describe the kinetics and the topology of grain growth and is valuable for theoretical research of microstructure evolution. However, there are some limitations existed in the algorithm which is being widely used presently,influencing the accuracy and efficiency of the grain growth simulation. It should be noted that if the simulation could not achieve the steady state of normal grain growth due to some limitations in the algorithm, both the kinetics of grain growth and the grain sign distribution functions obtained are not representative. On the basis of analyzing the existing MC algorithm, a modified MC method is proposed in order to study comprehensively the base and the meaning of the Hillert model of normal grain growth.

1 The Hillert Model of Normal Grain Growth

Drawing inspiration from Lifshitz and Slyozov's theory of particle coalescence[2], Hillert

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considered that the net increase in size of a grain(dR/dt) could be written as:

$$\frac{dR}{dt} = M\sigma g(\frac{1}{\rho_1} + \frac{1}{\rho_2})_{\text{average}} \quad (1)$$

where M is the mobility of the grain boundary, σ is the boundary tension, ρ_1 and ρ_2 are the principal radii of curvature, and g is a factor depending upon the shape of the grain. Since"an expression of the correct dimension and with the characteristic feature that it is positive for large R but negative for small R[2] is required, Hillert took"the simplest choice"[2] as:

$$g(\frac{1}{\rho_1} + \frac{1}{\rho_2})_{\text{average}} = \alpha(\frac{1}{R_C} + \frac{1}{R}) \quad (2)$$

where α is a constant of 1/2 and 1 in two and three dimensional systems respectively, R_C is the critical size or regarded as the average grain size of the system($<R>$). Combining equations (1) and (2) the average growth rate of all grains of size R is obtained:

$$\frac{dR}{dt} = \alpha M\sigma(\frac{1}{R_C} + \frac{1}{R}) \quad (3)$$

This is the well known Hillert's grain growth rate equation(usually noted as Hillert's basic equation), on the basis of which Hillert deduced the Grain Size Distribution Function(GSDF) of normal grain growth:

$$F(u) = (2e)^{\beta}\frac{\beta u}{(2-u)^{2+\beta}}\exp(-\frac{2\beta}{2-u}) \quad (4)$$

where β is the dimensions, and u is the relative grain size, $R/<R>$, in the two-dimensional case. Up to now, neither the experimental data nor the simulation results have been found to be consistent with the Hillert distribution, which is characterized by the right skewness and the maximum grain size, $R_{max}=2<R>$.

2 A Modified MC Method

The principal procedure of the existing MC method to simulate the normal grain growth is as follows[3 ~ 5] : a site is randomly selected and signed (or generally regarded as "orientated") with a value randomly selected among Q positive integers, a grain is constructed with neighbouring sites of the same orientation, and the grain boundary exists between sites of the different orientations. The probability to change the site orientation is determined by the energy change due to the change of the orientations configuration of sites. As far as the micro physical base of 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 in the simulation is proportional to the efficient reorientation probability of the sites. In the existing method, the reorientation attempt of a site is one of the possible orientation states of the whole system, the equivalent mobility rate decreases with the decrease of the efficient reorientation probability of the sites, thus in the later simulation period of grain growth the time exponent n will decrease, therefore, even under the infinite simulation time the time exponent of grain growth obtained from the existing MC method still deviates from the theoretical value $n=0.5$[9,10] . We spent a large amount of time tc investigate the grain growth kinetics with the existing MC method, but have never got any time exponent more than 0.43. This problem was also pointed out by Mulheran and Harding in their two and three dimensional grain growth simulations[7] . Hence the existing MC method is not suitable to simulate the total process of normal grain growth due to its limitations in the algorithm.

In order to modify the MC method, three points have been taken into consideration by the present authors. The first, according to the physical process of the grain boundary motion, the reorientation state of a site is determined as the same as one of its nearest neighbouring sites in the modified method and is no more related to the total number of the orientation states — the Q value. The second, a principle of "site selecting and reorientating one by one" is used in each cycling step, named as a MCS, instead of "site selecting randomly" in the existing methods[3 ~ 6] , which has not a reasonable meaning but increases the CPU time of computer in vain. The third, concerning the probability of site reorientation, if there is no energy change caused by the site reorientation, the site can change the orientation state or keep the original state with the same probability. The above three aspects leads to the modified MC method in present paper, which can be applied in the two dimensional simulations as well as three dimensnioal ones.

3 Simulation Results

With 200 × 200 micro-units selected to form a hexagonal network as the initial microstructure, the two-dimensional normal grain growth was simulated with the modified MC method.

3.1 The simulated time exponent of normal grain growth

Fig. 1 shows the variation of the average grain radius with the simulation time, the time exponents at different periods labellled. In the later period of simulated grain growth which is more close to the steady state, the time exponent obtained from the presen method is n=0.49 ± 0.01 and keeps almost unchanged later on.

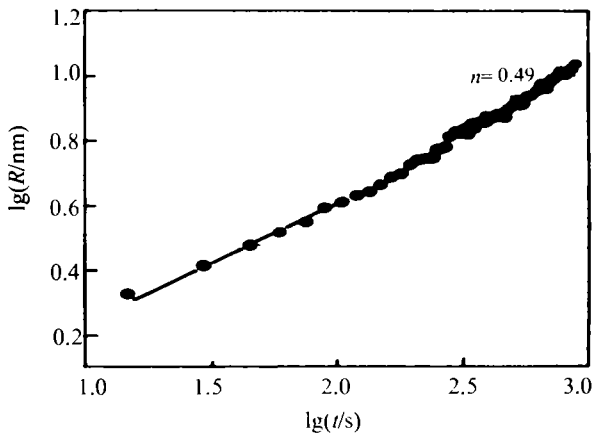

Fig. 1 Variation of the average grain radius with the simulated time, obtained from the present method

From the simulation results of grain growth kinetics, it can be seen that the modified MC method, basing on the physical process of grain boundary motion, can describe the total process of normal grain growth, including the grain growth of steady state; while the previous MC method, with complete randomness of site selection and reorientation, made the simulation intrinsically unable to achieve the steady state of normal grain growth (this can be seen from the poor simulated time exponent, which is much lower than the theoretical value n=0.5), consequently, the corresponding simulation results can't express the characteristics of steady grain growth precisely, but demonstrate those of a certain period of normal grain growth.

3.2 The Hillert and the von Neumann equations

Fig. 2 shows the plot of the average growth rate of grains within a size group of R_i vs the grain size, expressed in the form of $(1/<R>-1/R_i)$, where R_i is the equivalent radius of grain area, and $<R>$ is the average grain size of the system at the corresponding moment. Regressing several hundred pairs of the concrete data, a linear relationship is obtained as:

$$\frac{dR_i}{dt} = 0.13\left(\frac{1}{<R>} - \frac{1}{R_i}\right) \quad (5)$$

Comparing with equation (3), one can get the value of $M\sigma$ from the regressed scope tha $M\sigma$=0.26 in two dimensions. Fig. 3 displays, in another way, the average area variation rate of grains within a topological group as a functon of the topological parameter — the grain side number n, expressed in the form of $(n$-6). Regress from the same amount of concrete data as those in Fig. 3, anotiier linear relationship is obtained, equation(6).

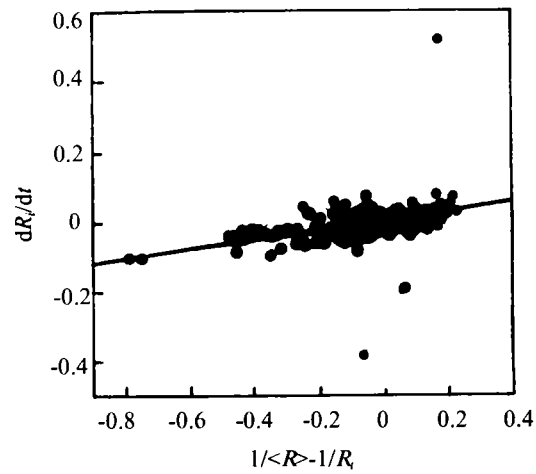

Fig.2 Variation of the average growth rate of grains in size group with R_i and the size expressed in the form of $(1/<R>-1/R_i)$

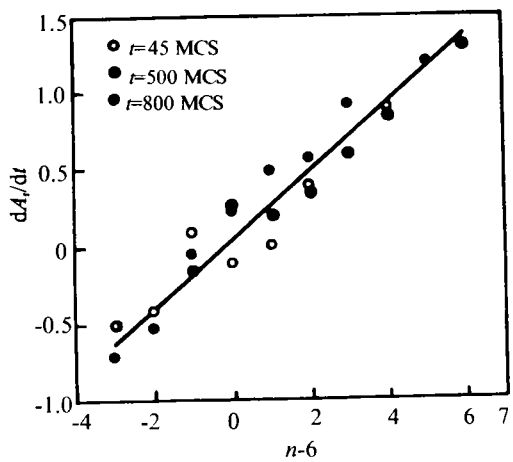

Fig.3 Average are variation rate of grains as a funcotin of the side number n, expressed in the form of $(n$-6), at different simulated time

$$\frac{dA_i}{dt} = 0.2303(n-6) \quad (6)$$

where A_i is the area of the grain with n sides. From the von Neumann equation[11]:

$$\frac{dA_i}{dt} = \frac{\pi}{3} M\sigma(n-6) \quad (7)$$

we can calculate the value of $M\sigma$ as $M\sigma$=0.22, which is very close to the value obtained from the regression of Hillert equation. In his original paper[2], Hillert applied the result of von Neumann and Mullins in order to determine the value of α, and realized that if his basic equation (equation(3)) was consistent with von Neumann's equation (equation(5)), the relation should hold between the size R and the average number of grains of that size in two dimensions:

$$n = 6 + 6\alpha\left(\frac{R}{R_C} - 1\right) \quad (8)$$

To further study the above relationship which connected the Hillert and the von Neumann equations, the interelation between the grain side number n and the grain size R was investigated at different simulated time with the present method. The results are shown in Fig. 4. It can be observed that the average side number n_R and the relative grain size $R/<R>$ obey the linear relationship approximately invariant during the long time simulation, which can be regressed as:

$$n_R = 3.01 + 2.82\frac{R}{<R>} \quad (9)$$

which is basically consistent with the equation (8) in the two-dimensional system of α=1/2. Therefore, the Hillert and the von Neumann equations are identical essencially according to the topological interelation. Since the von Neumann equation was deduced on the basis of the pure mathematical differentiation other than any assumptions of the grain shape and the grain boundary curvature[11], it is generally regarded as a strict equation to identify other theoretical models. However, the von Neumann model is in fact a geometrical model and appliable only in two dimensions. The Hillert model, considering the growth feature of grains within a certain size group, is more meaningful statistically and more practical both in two and three dimensional systems. Moreover, Hillert model has a clear physical base with consideration of the growth driving force of the grain boundary curvature.

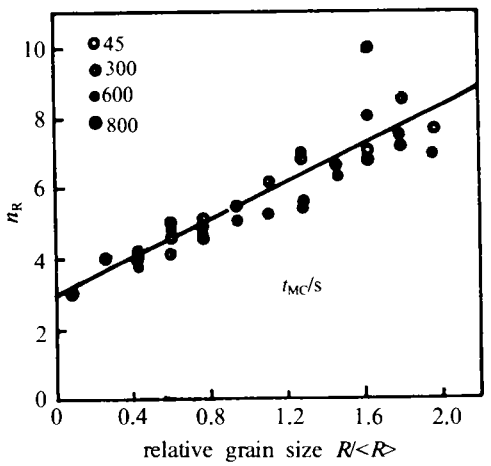

Fig.4 Relationship between the average side number of grains with size R to the relative size $R/<R>$, at different simulated time

3.3 Grain size distribution

The distribution of the relative radius of equivalent grain area during the simulated stage of t(MC) ≤ 500 s(where 500 s correspondings to lg$t$=2.7 when the simulated n first reaches 0.49), can be expressed properly by the lognormal functions with a little different parameters, as indicated in Fig. 5, which displayed the "time-invariability"[5], or sc called"self-similarity". As the grain growth proceeds, the GSD deviated from the former shape. Fig. 6 shows the histogram at time t_{MC} = 800 s, togethe: with the lognormal and the Hillert distribution of this moment. It is easy to find that the Hillert function showed better agreement with the at t_{MC}=800 s GSD than the lognormal function, and this fact maintained theretafter a period of 100 s or so. As

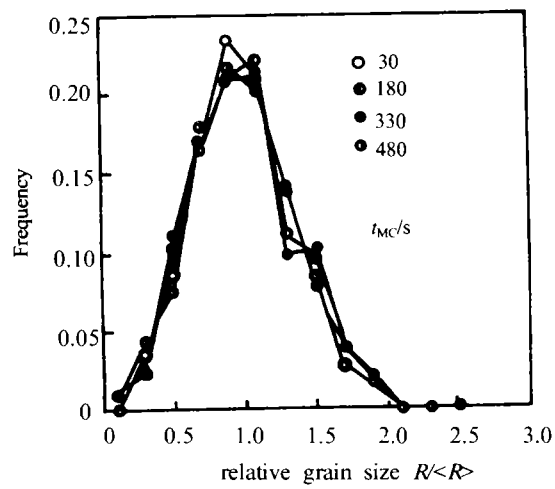

Fig.5 GSDs during simulated time t_{MC} ≤ 500 s, which can be expressed properly by the lognormal functions with a little different parameters

the simulated time exceeded 900 s, the grain number has decreased to fewer than 50, which made the statistics less convincing.

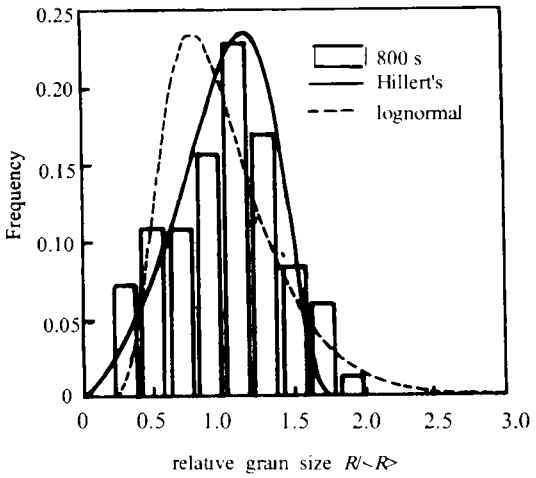

Fig.6 Histogram of GSD at simulated time t_{MC}= 800 s, compared with the lognormal and Hillert distributions

The above simulation results showed that the GSD does not keep an unique shape if the simu-lated time is sufficiently long, but varies continuously and slowly with the time in the process of grain growth. When the normal grain growth achieves the steady state where n is close to 0.50, the corresponding GSD reaches the form of the Hillert distribution.

4 Discussions and Conclusions

It is generally considered that the invariability of GSD is one of the characteristics of normal grain growth[5], and the invariant GSD has been observed in some experiments[12 ~ 14] and obtained from some computer simulations[3 ~ 6]. 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 stage of normal grain growth. Although the researched time scope of computer simulation was greatly improved comparing with the experimental investigation, the steady state of grain growth may not be attained due to the limitations in the algorithm. As a result, the so called time-invariant GSD, both from the experiments and the uncomplete simulations, can't represent GSD of the complete normal grain growth. Since the GSD with the shape quite similar to the lognormal function usually keeps a long time with a little change in the variation coefficient, the lognormal distribution is always taken as the "self-similar" distribution of normal grain growth both in experiments and some simulations, but it is not the distribution form of the steady grain growth where $n \approx 0.50$. This can be understood from the variation of the time exponent in n Fig. 1. The GSD of the quasi-steady state, corresponding to the time exponent infinitely close to the theoretical value n=0.50, has the form approximately consistent with the Hillert function(see Fig. 6). This means that the Hillert distribution does exist and it is one generated during the GSD forms of the total grain growth process, seemingly corresponding to the 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 impossible to keep the polycrystalline materials performing the normal grain growth unlimitedly in practice; the other is that the previous MC simulations restrain the grain growth to achieve the steady state, e. g. the simulated time exponent is never greater than n=0.43, which is muc smaller than the theoretical value.

The presently modified MC method allowed the study of the total process of normal grain growth, and it was found that the lognormal and the Hillert functions are the two expression forms of GSD during its transition with time, and the later seemingly corresponds to the steady state.

From the simulation results and discussions above, we draw conclusions as follows:

1) On the basis of analyzing the limitations in the existing MC algorithms, a modified MC method was proposed to simulate the normal grain growth in two dimensions, and the simulated time exponent of grain growth at quasi-steady state is very close to the theoretical value n=0.5.

2) According to the simulation results, the Hillert and the von Neumann equations are statistically identical, their connection of the topological interelation is identified. Hillert's basic equation appears to hold during the process of normal grain growth.

3) The grain size distribution generated during the complete grain growth process was found to vary continuously and slowly with the simulated time, the Hillert function seemingly corresponds to

the distribution at the steady stage where $n=0.50$.

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