Monte Carlo Simulation for the Behavior of Low Mobility Grain Boundaries in Grain Growth*

Toshiya Shibayanagi**, Hideaki Takatani*** and Shigenori Hori****

The change in frequency of grain boundaries with low mobility was investigated during grain growth using the Monte Carlo simulation. 4900 cells were generated and each grain was consisted of several cells in the initial condition. Each grain was randomly given a real number which corresponds to a grain orientation. Two types of grain boundaries classified for a mobility were characterized by the difference of each grain's number. The mobility ratios were 1:1, 1:2, 1:10 or 1:100 and the fractions of grain boundary with low mobility in the initial state were 8, 32 and 73%.

Grain boundaries with lower mobility tended to increase their existence ratio during grain growth, and this tendency became more noticeable with increasing mobility ratio. The frequency of grain boundary with lower mobility increased from 32% at the initial state to 55% as the grain size increased by four times. The drastic change of the fraction was observed when the grain size became large enough compared to the specimen area.

From the initial state of 73%, the frequency of grain boundary with lower mobility increased up to 83%. However, little change of the frequency was observed in the case with the initial state of 8%. Meanwhile, there existed some areas of localized grain boundaries with lower mobility and the change of microstructure in these areas was less than those of the other areas.

Thus, the controlling factors for changing the fraction of the grain boundaries with lower mobility are; (1) the initial fraction of grain boundaries with lower motility, (2) ratios of the mobilities and (3) the distribution of the boundaries with lower mobility.

(Received August 15, 1990)

Keywords: grain boundary character, grain growth, Monte Carlo simulation, grain boundary mobility ratio, lower mobility grain boundary

I. Introduction

There are many reports on the role of a grain boundary in the characteristics of materials. It has been made clear that a grain boundary with higher regularity of atomic arrangement such as the coincidence boundary are difficult sites for the grain boundary failure, the discontinuous precipitation or the stress corrosion cracking⁽¹⁾⁻⁽⁶⁾ and so on. Moreover, the relationship between these phenomena and the grain boundary characters is also affected by deformation conditions or aging conditions⁽⁷⁾⁽⁸⁾. These results suggest that the improvement of the characteristics of material is expected by making use of the grain boundary character in itself besides the conventional techniques of controlling microstructures such as an addition of third elements⁽⁹⁾⁻⁽¹¹⁾.

Recently, a new material design using these grain boundary characters has been proposed⁽¹²⁾, and the researches are now in progress to realize it. As many production processes of materials contain grain growth, we have to understand first the change of the distribution of the grain boundary character in the grain growth. There

are many reports on the control of grain orientations in Fe-Si alloys or the textures⁽¹³⁾⁽¹⁴⁾, while there are only a few reports concerning the distribution of the grain boundary character.

Watanabe *et al.*⁽¹⁵⁾ found an increase of the frequency of the coincidence boundaries in an annealed Fe-6.5%Si alloy ribbon which was rapidly quenched. They also showed an improvement of ductility of the alloy with increase of the frequency of coincidence boundaries. Shimizu and Harase⁽¹⁶⁾ reported a role of coincidence boundaries in the development of the gross texture.

These results suggest the possibility of the improvement of the characteristics of materials by controlling the distribution of grain boundary character in the grain growth. However, they also observed the combined effects of some factors such as solute atom, precipitates, annealing temperature and so on.

Thus, a computer simulation for the grain growth using some known factors is effective to reveal the controlling factors which change the distribution of grain boundary character in grain growth.

In the present study, the Monte Carlo simulation was employed to simulate the grain growth, and the change of the frequency of grain boundaries with low mobility was estimated.

II. Experimental Procedure

1. A polycrystalline model

4900 hexagonal cells were generated and a grain consists of several cells. Each grain has a random real

* This paper was originally published in Japanese in J. Japan Inst. Metals, **54** (1990), 650.

** Department of Materials Science and Engineering, Faculty of Engineering, Osaka University, Suita, Osaka 565, Japan.

*** Graduate Student, Osaka University, Osaka. Present address: Mitsubishi Heavy Industries, Ltd., Hiroshima Research & Development Center, Hiroshima 733, Japan.

**** Department of Materials Science and Engineering, Faculty of Engineering, Osaka University, Osaka. Present address Teikyo University, Utsunomiya, Tochigi 320, Japan.

numeral ranging from 0 to N ($N=40, 90$ and 360), corresponding to a grain orientation. Grain boundaries are characterized by the difference of the numerals of two grains, and the difference corresponds to the misorientation in real materials. Then, the grain boundaries are classified into two types in the following manner.

If the difference of the numerals of two grains is less than a critical value, the grain boundary is called the special boundary and another type of a boundary with the difference larger than the critical value is called the random boundary. In the present work, the critical value was fixed to 15. This critical value of 15 corresponds to the allowance of the deviation angle from the exact orientation relation in $\Sigma 1$ boundary, which was proposed by Brandon⁽¹⁷⁾.

Meanwhile, the grain boundary mobilities are classified into two kinds depending on the grain boundary characters where the grain boundary with the numeral difference less than 15 was called lower mobility-boundary and the boundary with the numeral difference larger than 15 was called higher mobility boundary.

The frequency of the grain boundary with lower mobility was changed by changing the number N , with the critical value fixed to 15.

2. Simulation of the grain growth

Grain growth was simulated using the algorithm presented by Anderson *et al.*⁽¹⁸⁾⁽¹⁹⁾ Figure 1 shows the cell model of grain boundary migration. Each cell has a numeral corresponding to an orientation and the numerals are denoted as A and B in the figure. If the energy of each cell is denoted as E , then the value is defined as:

$$E = \Sigma(1 - \delta n_i n_j) \quad (1)$$

where n_i, n_j mean the numerals representing the orientation, and i and j mean the cell number. δ is Kronecker's delta.

The contribution of a cell to the energy state of its surrounding region can be estimated by eq. (1).

Grain boundary migration was simulated as follows. First, one cell is randomly selected in the simulating area of 4900 cells. For example, the cell enclosed with a circle is chosen and then another adjacent cell is selected. If the change of E is negative when the numerals of the two cells are exchanged, the region changes to more stable

state and the segment of the grain boundary can migrate. Hence, the grain boundary migration is performed by replacing the numeral A by the numeral B, as shown in Fig. 1(b). This model represents that the grain boundary migration occurs to reduce the total area of grain boundary in a unit volume.

Secondly, a new numeral is randomly given for this segment of boundary which satisfy the above moving condition. Then, the probability of grain boundary migration can be settled, assuming that the segment of grain boundary is allowed to move if this new numeral is less than another fixed value. Therefore, this probability of migration can be taken as the mobility of grain boundary in this paper.

In the case of two kinds of mobility, the probability of the migration for the grain boundary with higher mobility was set as unity. The probability of the migration for the grain boundary with lower mobility was changed from 0 to 1, such as 0.1 or 0.5.

In the present study, the ratios of the two mobilities were set to be 1:1, 1:2, 1:10 and 1:100. The calculation of 4900 times of this procedure was defined as 1 MCS (Monte Carlo step), which means that all the cells were selected at least one time. The grain structure obtained after the 10 MCS of computation with the single kind of the mobility was adopted as the initial state and there observed no island grain.

III. Results

1. Grain growth with single mobility

Figure 2 shows an evolution of the grain structure during grain growth. The broad grain boundary represents the grain boundary whose difference of the numeral is less than 15. Every grain boundary has the same mobility regardless of the grain boundary character. No abnormal large grain existed at each step. Hence the changes of the structures simulates a normal grain growth. There were some regions where a contact of two grain boundaries generated a new orientation relationship or the generation of a new kind of grain boundary character.

Figure 3 shows the relationship between the grain size and the Monte Carlo step for the single kind of mobility. There was a linear relation with the slope of 0.38. This value was in good agreement with that reported by Burke and Turnbull⁽²⁰⁾ in the normal grain growth. Thus, the MCS in this study can be taken as the real annealing time.

Figure 4 shows the change of the frequency of the special boundary during the grain growth. R and R_0 in the abscissa denote the mean grain diameter of each step and that of the initial state, respectively. The frequency of the special boundary at the initial state was 35% and the frequency changed between 26 and 40% in the following grain growth. Thus, the frequency change is very small when a single mobility of grain boundary is assumed independent of the grain boundary character.

Fig. 1 Two dimensional cell model of grain boundary migration by the Monte Carlo simulation.

Fig. 2 An evolution of microstructure during grain growth. The mobility ratio is 1:1 and the initial fraction of special boundaries is 35%.

Fig. 3 Relationship between the average grain size (R) and the Monte Carlo step (MCS).

Fig. 5 Change in the fraction of grain boundaries with lower mobility during grain growth. The mobility ratios are 1:2, 1:10 and 1:100 and the initial fraction of grain boundaries with lower mobility is 32%.

Fig. 4 Change in the fraction of special boundaries during grain growth. The mobility ratio is 1:1 and the initial fraction of special boundaries is 35%.

2. Grain growth with two kinds of mobility

Grain growth with two kinds of mobility classified by the grain boundary character was simulated as shown from Fig. 5 to Fig. 8.

Figure 5 shows the change of the frequency of the grain boundary with lower mobility. The initial frequency was 32%. The frequency of the grain boundary with lower mobility increased until the grain size became four times as large as the initial one. When the difference of the two mobilities became larger, the increment of the frequency of the lower mobility boundary increased. For example, when the mobility ratio was 1:100, the frequency increased up to 58% which was twice the initial frequency. When the grain size became four times larger than that of the initial state, the frequency decreased less than that of the initial state in the case of the

Fig. 6 An evolution of microstructure during grain growth. The mobility ratio is 1:100 and the initial fraction of lower mobility boundaries is 32%.

Fig. 7 Change in the fraction of grain boundaries with lower mobility during grain growth. The mobility ratios are 1:2, 1:10 and 1:100 and the initial fraction of grain boundaries with lower mobility is 73%.

Fig. 8 Change in the fraction of grain boundaries with lower mobility during grain growth. The mobility ratios are 1:2, 1:10 and 1:100 and the initial fraction of grain boundaries with lower mobility is 8%.

mobility ratios of 1:2 and 1:10. On the other hand, the frequency of the lower mobility boundary increased up to 60% for the mobility ratio of 1:100 when the grain size became six times larger than the initial one.

Figure 6 shows the grain structures at each step in Fig. 5. The broad grain boundary represents the lower mobility boundary. The region enclosed with a circle has more grain boundaries with lower mobility than that in the other area. The grain structure in this area was stable until the final state of the grain growth.

The contact of grain boundaries or the extinction of grains formed new grain boundaries and the characters of these grain boundaries were not always the same as those existing before the extinction. In addition, there were some areas where the distribution of the grain

boundary character changed locally. These local changes of the distribution had some effects on the successive change of the grain structure.

Figure 7 shows the change of the frequency of the grain boundary with lower mobility when the initial frequency was 73%. As for the mobility ratio of 1:100, similar to Fig. 5, the frequency increased with grain growth and it became about 90% when the grain grew twice as large. Meanwhile, as the grain growth proceeded, the frequency of low mobility boundary tended to decrease for every mobility ratio.

When the initial frequency was 8%, as shown in Fig. 8, no apparent change was observed until the grain size increased four times as large as the initial one for every grain boundary mobility ratio. However, the frequency

changed drastically in the latter state and the value was about 20% when the grain size was eight times as large as the initial grain size.

IV. Discussion

1. Grain growth model

The grain growth of real materials is complicated functions of many kinds of mobility. The grain boundary migration depends on many factors as follows; temperature, grain boundary structure, segregation of solute atom or impurities, pinning by precipitates or second phase particles, degree of working, elastic stress, surface structure, oxide film, atmosphere and so on. The grain boundary migration is affected by a single factor or by some combined factors.

Grain boundary was characterized with one or two kinds in the present model. Two kinds of grain boundary character simulates the grain growth where small angle grain boundaries have different mobility from those of other grain boundaries. For example, an annealing at high temperature or the development of a texture can be simulated by this model.

The ratio of the mobilities is also functions of the above factors and the grain growth reflect these combined effects.

In the present study, as shown in Figs. 5, 7 and 8, a salient effect appeared for the mobility ratio larger than 1:10. Moreover, if the grain became large as compared with the simulating area, the coalescence of grains hardly occurred in the edge region. Hence, the change of grain structure in the edge region differs from that in the central area, simulating the structural change of the thin foil materials or the near-surface region of the bulk materials.

2. Role of grain boundary characters in grain growth

If the mobility of grain boundary depend on the grain boundary characters, the frequency of the special boundaries and the lower mobility boundary increased with increase of grain size, as shown from Fig. 5 to Fig. 8. From these results, it was made clear that the mobility of grain boundary and the ratio of the mobilities play an important role in the grain growth.

The frequency of the lower mobility boundaries (α_i) at each step is defined as:

$$\alpha_i = S_i / (S_i + R_i) \quad (2)$$

where S_i and R_i are the numbers of the lower mobility boundaries and random boundary at the i th step, respectively.

As seen from Fig. 5 to Fig. 8, α_0 is an important factor in the grain growth because α_i increased when α_0 increased. In addition, there was a critical value of α_{crit} where a drastic increase of α_i occurred. α_{crit} lied between 8% and 32%.

α_0 must be larger than α_{crit} in order to increase the lower mobility boundaries.

By the way, a new orientation relation is made successively by a contact of two separated grain boundaries at each state of grain growth. Hence the probability of the birth of the lower mobility grain boundary is required to be high in order to increase the frequency of the lower mobility grain boundary. The reaction of forming a new grain boundary character in grain growth is represented as:

$$\Sigma_{ab} + \Sigma_{bc} = \Sigma_{ac} \quad (3)$$

where, Σ_{ij} means a character of grain boundary between a grain i and a grain j . A different kind of grain boundary can be formed in some combinations of grains at each stage in the grain growth.

Therefore, at the initial state or at each step of grain growth, the change of the frequency depends on the behavior of the “imaginary” grain boundary which will be formed in future as well as the existing grain boundary. In the simulation shown from Fig. 5 to Fig. 8, α_0 was changed by limiting the maximum value of N . Thus, the large value of α_0 means the large frequency of the imaginary grain boundary with lower mobility, suggesting the importance of the role of the character of the imaginary grain boundary.

On the other hand, as shown in Fig. 6, the distribution of the grain boundary character differed locally from each other and these fluctuations had effects on the change of structure in grain growth. Hence, the change of the frequency of the lower mobility boundary is not determined only by the frequency of the real or the imaginary boundaries with lower mobility. Therefore, the spacial distribution of the location of the grains must be taken into account for the formation of new grain boundaries given by eq. (3).

Grain growth can be regarded as the extinction process of grains, and an example of the process is shown in Fig. 9. Five grain boundaries disappears by the extinction of the grain 0. In the process of the extinction, as shown from (b) to (d), two new orientation relations of 1-4 and 2-4 are formed from the grain boundaries of 1-0 and 4-0 and those of 2-0 and 4-0, respectively. If the grain boundary is characterized as “random” and “special”, the formation of a new grain boundary by a contact of two grains is classified into the following six equations which are the expanded types of eq. (3);

$$\begin{aligned} \text{random} + \text{random} &= \text{special} \\ \text{random} + \text{special} &= \text{special} \\ \text{special} + \text{special} &= \text{special} \\ \text{random} + \text{random} &= \text{random} \\ \text{random} + \text{special} &= \text{random} \\ \text{special} + \text{special} &= \text{random} \end{aligned} \quad (4)$$

If every grain boundary has the same mobility, the frequency of the special boundary unchanged during grain growth as shown in Fig. 4. In this case, the frequency of the special boundary at each step remained the same value as the initial frequency after the extinction and the

Fig. 9 Change of grain boundary character by an extinction of a grain.

formation of grain boundaries. However, in the case of two mobilities of grain boundary, as shown in Fig. 5, the frequency of grain boundary with lower mobility increased with grain growth. These results suggest that the balance of eq. (4) was broken.

Therefore, characters of grain boundaries belonging to the extinguishing grain and the character of newly formed grain boundary are important in the change of the distribution of the grain boundary characters in grain growth. The frequency of lower mobility grain boundary increases if there are higher mobility grain boundaries around the extinguishing grains or the newly formed grain boundaries have lower mobility. Thus, the important factor determining the distribution of the grain boundary character at each step in the grain growth is the distribution of grains whose orientations make the low mobility grain boundary.

Meanwhile, grain boundary mobilities are also the important factor in the extinction of grains. The frequency of the grain boundary with lower mobility increased rela-

tively because the life of grains containing more grain boundaries with lower mobility was longer than the other grains.

In other words, the formation of new grain boundaries is a function of the grain boundary mobility. Hence, the probability of the formation of grain boundary with lower mobility depends on the kind of grain boundary of the extinguishing grains and the orientation of grains around the extinguishing grains.

Change of the distribution of the grain boundary characters in the edge area is less than that in the central area because the probability for the contact of grains is small in the edge area. Moreover, grain boundaries with higher mobility have higher probability for getting out the simulating area than those with lower mobility. Consequently, the frequency of lower mobility grain boundaries increased.

The effect of grain boundary mobility became apparent when the difference of the two mobilities increased, as shown from Fig. 5 to Fig. 8. These effects appear in the grain growth of thin foils or the edge region of bulk materials.

Therefore, we can control the distribution of the grain boundary characters by controlling the frequency, the mobility ratio or the spacial distribution of grain boundaries with different mobilities.

V. Conclusions

Monte Carlo simulation of grain growth was performed to investigate the changes of the frequency and the distribution of the special grain boundaries or the low mobility grain boundaries during grain growth with a single or two kinds of mobilities.

As for two kinds of mobility, the simulation was performed with various ratios of grain boundary mobility and with the variation in the initial frequency of grain boundary with lower mobility.

The following results were obtained.

(1) The frequency of the special boundaries changed slightly during the grain growth if every grain boundary had the same mobility.

(2) The frequency of low mobility boundaries tended to increase with increase of a grain size and the increment of the frequency increased when the difference of the two mobilities increased. However, the change of the frequency of low mobility boundary was small when the initial frequency of the grain boundary with lower mobility was 8%.

(3) There was a drastic change of the frequency of the special boundary or the low mobility boundary when the grain size became large enough as compared with the simulating area.

(4) The change of the ratio of the lower mobility boundary was not monotonic and was affected by the orientation distribution or the spacial distribution of grain boundary characters at each stage of the grain growth.

Acknowledgment

The present work was financially supported by the Grant-in-Aid of The Ministry of Education, Science and Culture, Japan (No. 62420047 and No. 01750707).

REFERENCES

- (1) T. Watanabe: Met. Trans., **14A** (1983), 531.
- (2) L. C. Lim and R. Raj: Acta Metall., **32** (1984), 1183.
- (3) M. Frebel and J. Schenk: Z. Metallk., **70** (1979), 230.
- (4) W. Gruhl and D. Amman: Acta Metall., **3** (1955), 347.
- (5) M. H. Ainsley, G. J. Cocks and D. R. Miller: Met. Sci., **13** (1979), 20.
- (6) T. Asahi, F. Yabusaki, K. Osamura and Y. Murakami: Proc. 6th Int. Conf. on Light Metals., (1975), p. 64.
- (7) T. Shibayanagi, A. Yoshimoto and S. Hori: Mater. Trans., JIM, **31** (1990), 1057.
- (8) T. Shibayanagi, Y. Kitazume and S. Hori: J. Japan Inst. Metals, **53** (1990), 9.
- (9) W. Gruhl and G. Wassermann: Metall., **5** (1951), 141.
- (10) N. Igata, S. Sato, T. Morimoto and T. Ogura: J. Japan Copper and Brass Research Association, **17** (1978), 156.
- (11) M. Omori, K. Wakasa and H. Yoshinaga: J. Japan Inst. Metals, **37** (1973), 1188.
- (12) T. Watanabe: *Interfacial Structure, Properties and Design*, M. H. Yoo *et al.* ed., MRS, (1988), p. 443.
- (13) D. J. Jensen, N. Hansen and F. J. Humphreys: Acta Metall., **33** (1985), 2155.
- (14) J. E. May and D. Turnbull: Trans. Met. Soc. AIME, **212** (1958), 769.
- (15) T. Watanabe, H. Fujii, H. Oikawa and K. I. Arai: Acta Metall., **37** (1989), 941.
- (16) R. Shimizu and J. Harase: Acta Metall., **37** (1989), 1241.
- (17) D. G. Brandon: Acta Metall., **14** (1966), 1479.
- (18) M. P. Anderson, D. J. Srolovitz, G. S. Grest and P. S. Sahni: Acta Metall., **32** (1984), 783.
- (19) A. D. Rollett, D. J. Srolovitz and M. P. Anderson: Acta Metall., **37** (1989), 1227.
- (20) J. E. Burke and D. Turnbull: Prog. Metal Phys., **3** (1952), 220.