Analysis of Growth Behavior of a Cellular and Dendritic Interface under a Constrained Growth Condition using a Phase-Field Model

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A phase-field simulation was carried out to investigate the growth behavior of a cellular and dendritic interface of an Fe–C binary alloy in a constrained growth condition. The simulated results were in good agreement with the experimental results. The effect of the magnitude of anisotropy at solid/liquid interface energy was examined, and it was found that the magnitude of anisotropy affected the growth direction when the growth rate was low. Dimensionless growth direction, \( \frac{V}{V_c} \), was used to examine the obtained results, and it was found that \( \frac{V}{V_c} \) increases from zero and approaches unity with increase in growth velocity. A good correlation was obtained between calculated growth velocity and growth direction by using dimensionless growth velocity (\( V/V_c \)), and this correlation was in agreement with the experimental results.

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1. Introduction

The mechanism of ‘grain selection’ plays an important role in fabrication of single crystal products using a unidirectional solidification technique such as turbine blades.1) Although single crystal products have been skillfully made by using grain selection, the mechanism of the grain selection has not been elucidated, and a trial and error method is required to make these products. To understand the mechanism of grain selection, the relation between growth direction of cells or dendrites and solidification condition should be elucidated.

It is well known that the growth direction of cells or dendrites is determined by the preferred growth direction and heat flow in a constrained growth condition. Kurz and Fisher2) described the growth directions of dendrites and cells found in normal cubic metals, the preferred growth direction of which is \( \langle 100 \rangle \), in the case of constrained growth. Columnar dendrites grow in their preferred growth direction, \( \langle 100 \rangle \), regardless of the direction of heat flow. On the other hand, cells grow in the heat flow direction regardless of their preferred growth direction. However, they did not give the reason for the change in growth direction during the transition from cells to dendrites.

In our previous work,3) in situ observation using succinonitrile (SCN) was performed, and the relation between growth velocity and growth direction of cells and dendrites was obtained. In the experiments, effects of both heat flow and preferred growth direction on either growth of cells or dendrites were observed. In addition, simulations using a phase-field model of an Fe–C binary alloy were carried out to interpret the experimental results, and the simulated results qualitatively agreed with the experimental results. However, since phase-field calculation requires a long CPU time, simulation in previous study has been carried out under a non-steady state growth condition, whereas experiments have been carried out under a steady state condition. In addition, it is expected that the degree of anisotropy of materials influences the growth behavior of cells and dendrites; however, the effect of anisotropy has not been examined yet.

In the present study, the growth behavior of a cellular and dendritic interface in a constrained growth condition was simulated under a steady state condition using a phase-field model, and the results obtained were compared with experimental results. The effect of the magnitude of anisotropy on growth direction of cells and dendrites was also investigated.

2. Simulation Method

2.1 Phase-field model

In the present simulation, the phase-field model proposed by Kim et al.4,5) was used. Since the model has been described in detail in our previous paper,3) only a brief description of the model will be given here. The governing equations of the phase-field model for an alloy are as follows:

\[
\frac{\partial \phi}{\partial t} = -M(c^2 \nabla^2 \phi - f_c),
\]

\[
\frac{\partial c}{\partial t} = \nabla \cdot \left( \frac{D(\phi)}{f_{cc}} \nabla c \right),
\]

where \( f(c, \phi) \) is free energy density of a solid/liquid mixture, \( D(\phi) \) is the solute diffusion coefficient, \( M \) is a phase-field parameter related to kinetics, and \( \epsilon \) is a phase-field parameter related to interfacial energy and is expressed as

\[
\epsilon(\theta) = \epsilon(1 + \delta \cos 4\theta),
\]

where \( \theta \) is the angle between the normal of a solid/liquid interface and the x-axis, and \( \delta \) is the magnitude of anisotropy (called anisotropy parameter).

2.2 Calculation procedure

In the simulation, the calculation domain was divided into uniform square grids, and eqs. (1) and (2) were changed to explicit finite difference forms and numerically solved. An Fe–0.2 mass%C binary alloy was used as the target material, as it was in our previous study.3) The physical properties and thermodynamical data of the Fe–C binary alloy6) used in the present simulation are shown in Tables 1 and 2, respectively. The phase-field parameters used in the simulation were set as

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Table 1 Physical properties of Fe–C binary alloy.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diffusion coefficient of liquid, $D_L$ (m²/s)</td>
<td>$2.0 \times 10^{-8}$</td>
</tr>
<tr>
<td>Diffusion coefficient of solid, $D_S$ (m²/s)</td>
<td>$6.0 \times 10^{-9}$</td>
</tr>
<tr>
<td>Interface energy, $\sigma$ (J/m²)</td>
<td>0.204</td>
</tr>
<tr>
<td>Partition coefficient, $k_p$</td>
<td>0.196</td>
</tr>
<tr>
<td>Slope of liquidus line (K/mol fra.)</td>
<td>$-1932.9$</td>
</tr>
<tr>
<td>Melting point of Fe (K)</td>
<td>1810</td>
</tr>
</tbody>
</table>

Table 2 Thermodynamic data of δ crystal and liquid phase of Fe–C binary alloy.

$$
G = (1 - x)G_{Fe}^0 + xG_{C}^0 + [x \ln(x) + (1 - x)\ln(1 - x)] \\
+ x(1 - x)/[L_0 + (1 - 2x)] (J/mol)
$$

- $G_{Fe}^0 = 0$
- $G_{C}^0 = 146858 - 14.644T$
- $G_{Fe}^{391} = 13807.2 - 7.6316T$
- $G_{C}^{391} = 114223 - 27.196T$
- $L_{Fe}^{0} = -65480 - 33.451T$
- $L_{C}^{0} = 30334 - 4.372T$
- $L_{Fe}^{391} = -123846.5 + 29.7485T$
- $L_{C}^{391} = 32216.5 - 19.2045T$

The calculation domain was divided into $400 \times 2000$ square grids, each 0.02 μm in size. To simulate a cellular and dendritic interface, the initial condition shown in Fig. 1 was adopted. The boundary condition in the calculated domain was that the concentration gradient of the solute was zero. In addition, a time-saving calculation technique by which the calculation was not carried out for all grids but only for some grids in a certain region containing the solid/liquid interface was used to simulate the growth of cells or dendrites under a steady state condition. The calculation region in the growth direction (X-axis direction) contains hundreds of grids on either side of the interface. The calculation region was moved with change in the position of the solid/liquid interface. In the case of high growth velocity, 400 (the center being the solid/liquid interface) grids in the x-axis direction were selected for the calculation, and in the case of low growth velocity, 800 grids were selected because the diffusion boundary layer in front of the solid/liquid interface changes depending on the growth velocity. The calculated domain in a 2-dimensional condition consists of $400 \times 400$ and $800 \times 400$ grids. The calculation time for this domain was only 1/2 or 1/3 of that in the case of calculating all grids. To confirm the validity of the time-saving technique, the results of calculation were compared with the results obtained by calculating all of the grids. It was found that the coarsening process proceeds at the roots of dendrites and cells in the case of calculating all of the grids more than it dose in the case of using the time-saving technique. However, the same morphology and growth behavior of the tips of cells and dendrites were observed.

A certain positive temperature gradient was imposed in the calculation domain, and all of the grids were set to be solid at $x = 0$ as an initial condition. The preferred growth direction was the x-axis direction, and the tilted angle between heat flow and preferred growth direction was set to be about 18.43 degrees (= arctan(1/3) rad.). In the calculation, the heat transfer equation was not explicitly calculated, and the effect of temperature gradient was involved by changing the free energy densities and the phase-field parameter as a function of temperature. The temperature profile was moved in the heat flow direction at a certain constant velocity to emulate the real experimental conditions. A schematic illustration of the experimental apparatus using succinonitrile is shown in Fig. 2, and the experimental procedure has been described in detail in our previous paper. The simulated experimental conditions are shown in Table 3.

To investigate the effect of anisotropy, $\delta$, in eq. (3), three values of the anisotropy parameter, 0.03, 0.04 and 0.05, were examined. It is known that the anisotropy parameter in eq. (3) must be less than 1/15, and the three values of the anisotropy parameter were set to be less than 1/15. As a result, it was confirmed that dendrite morphology can be produced with the anisotropy parameter in the above range.
3. Results and Discussion

3.1 Results of calculation and comparison with experimental results

Figure 3 shows the calculated solid/liquid interface morphology and microsegregation patterns of the solute during solidification of the Fe–0.2 mass%C alloy in a steady state condition. The gray scale in each of the figures corresponds to the carbon content. Figures 3(a), (b), (c) and (d) show the calculated growth morphology of cellular and dendritic interfaces whose growth velocities are $9.5/10 \times C^2/10$ m/s, $3.8/10 \times C^2/10$ m/s, $1.0/10 \times C^2/10$ m/s and $1.9/10 \times C^2/10$ m/s, respectively. In the case of a cellular interface growing at a low velocity (see Figs. 3(a) and (b)), the growth direction deviates from the preferred growth direction to the heat flow direction. On the other hand, in the case of a cellular and dendritic interface growing at a high velocity (see Figs. 3(c) and (d)), the effect of heat flow on the growth direction is slight. The morphology of cell tips becomes sharp toward the preferred growth direction with increase in growth velocity. To estimate the degree of deviation, dimensionless growth direction, $\pi'$ defined in our previous paper $3)$ was used. Dimensionless growth direction, $\pi'$ is defined as follows:

$$\pi' = \alpha/\theta,$$

where $\alpha$ is the angle between heat flow direction and growth direction, and $\theta$ is the angle between heat flow direction and preferred growth direction. The growth direction was determined by tracking a growing cell or dendrite tip after steady state growth had been achieved.

The calculated relation between $\pi'$ and growth velocity at $G = 1.58 \times 10^6$ K/mm is shown in Fig. 4. Since the growth behavior in the period of initial solidification is not in a steady state, $\pi'$ was measured from the growth morphology in the later stage. The dimensionless growth direction, $\pi'$, increased with increase in growth velocity and approached unity at a higher value of growth velocity. Although it is difficult to make a direct comparison of the results of the calculation and experiment due to the difference between the experimental system and the calculation system used, the trend in calculated results agrees with that in the experimental results $3)$. Figure 5 shows the relationship between growth velocity and dimensionless growth direction, $\pi'$ at $G = 3.16 \times 10^6$ K/m. The value of $\pi'$ increases with increase in growth velocity,
like the results shown in Fig. 4, but the curve of $\pi'$ in Fig. 5 rises more gradually than that in the case of $G = 1.58 \times 10^8 \text{ K/m}$ shown in Fig. 4. These results also agree with the tendency obtained from the experimental results.\(^3\)

### 3.2 Dimensionless growth direction

Figure 6 shows the relationship between normalized growth velocity, $(V/V_C)$, and $\pi'$ obtained by an experiment using SCN.\(^3\) Here, $V_C$ is the critical velocity for breaking down a planar interface and is expressed as follows:

$$V_C = \frac{-kDG}{mC_0(1-k)},$$ \hspace{1cm} (5)

where $k$ is the equilibrium partition coefficient, $m$ is the slope of the liquidus line, and $D$ is the diffusion coefficient of solute in the liquid. In the experiment, $\pi'$ increased sharply with increase in $V/V_C$ until it reached approximately 0.8. When $\pi'$ exceeded 0.8, it increased gradually with increase in $V/V_C$ and approached unity. The experimental data obtained from three sets of temperature gradient are found to fall on a unique curve.

Figure 7 shows the relationship between $V/V_C$ and $\pi'$ obtained in this simulation. The value of $\pi'$ increased sharply with increase in $V/V_C$ and approached unity as was obtained in the experiment. The calculated data obtained from two sets of temperature gradient may be also found to fall on a unique curve. However, the results of calculation shown in Fig. 7 are not exactly the same as the experimental results shown in Fig. 6. The reason for this difference may be the different alloy systems and different solidification conditions used in the calculation and experiment.

The relation between $\pi'$ and $V$ may depend on the value of anisotropy of the material. Therefore, the effect of anisotropy was examined. The value of the anisotropy parameter, $\delta$, used in the simulation for which results are shown in Figs. 3 to 5 and 7 was 0.04.

### 3.3 Effect of anisotropy on growth direction

In general, the value of $\delta$ in eq. (3) was empirically determined so as to produce appropriate dendrite morphology,\(^8,9\) however, it is not clear how the anisotropy parameter can be concerned with the anisotropy of interfacial energy.\(^10,11\) In the present study,\(^7\) three values of $\delta$ (0.03, 0.04 and 0.05) were used and the effect of anisotropy on growth direction was examined.

Figure 8 shows the calculated solid/liquid interface morphology and microsegregation pattern of the solute during unidirectional growth of the Fe–0.2 mass% C alloy for different values of the anisotropy parameter, (a) 0.03 (b) 0.04 and (c) 0.05, at $G = 1.58 \times 10^8 \text{ K/m}$ and $V = 9.5 \times 10^{-3} \text{ m/s}$. As can be seen in the figure, the growth direction of cells is influenced by the magnitude of the anisotropy parameter, and the cellular interface deflects toward the heat flow direction with a large angle when the magnitude of anisotropy becomes small.
Figure 9 shows the relationship between growth velocity and dimensionless growth direction for different anisotropy parameter. The magnitude of anisotropy had a marked influence on the growth direction in the low growth velocity region. However, the effect of anisotropy in the high growth velocity region was small. A marked effect of the magnitude of anisotropy on growth direction was observed in the deflection behavior of dendrites due to fluid flow even in the case of high growth velocity. In addition to the condition without fluid flow, the reason for this may be the steady state condition used in the present study. In any case, the tendency for $\pi'$ to increase with increase in growth velocity is similar for all values of $\delta$ and agrees with the experimental results.

4. Conclusions

Phase-field simulation was carried out to investigate the growth behavior of a cellular and dendritic interface of an Fe–C binary alloy in a constrained growth condition. In the simulation, a steady state condition was set up during the growth of cellular and dendritic interface by using a calculation technique. Deviation from the preferred growth direction to the heat flow direction was observed in the case of a cellular interface, whereas the dendritic interface grew in its preferred growth direction. The results of simulation were in good agreement with the experimental results. The effect of magnitude of anisotropy at the solid/liquid interface used in the phase-field simulation was examined, and it was found that the magnitude of anisotropy has a marked effect on growth direction when the growth rate is low.

The results obtained from both the simulation and the experiment are summarized as follows:

1. Dimensionless growth direction, $\pi'$, increase from zero and approaches unity with increase in growth velocity.
2. By introducing the dimensionless growth velocity ($V/V_c$), a good correlation described with a unique line was obtained between growth velocity and growth direction.

REFERENCES