1. Introduction

Control of the solidification structures of castings is a useful method for the control of properties and quality of casting products. Numerical modeling for simulating solidification structure formation makes it possible to analyze the mechanism of the evolution of micro and macrostructures and can provide useful information for controlling the solidification structure. In the past two decades, various kinds of deterministic and stochastic models for predicting the evolution of micro and macrostructures of alloys during solidification have been developed. The phase-field (PF) method is a deterministic model for simulating the evolution of microstructures such as dendrites of pure substances or alloys.\(^1\)\(^-\)\(^3\) This method is a very powerful tool for simulating the evolution of a dendrite structure.\(^4\)\(^-\)\(^8\) However, a drawback of the PF method is that the use of the PF method for simulation of macrostructure formation in a casting such as equiaxed and columnar grains will be difficult.

Stochastic models such as a Monte Carlo (MC) method and a cellular automaton (CA) method have been used for simulating the evolution of grain macrostructures in solidifying alloys.\(^9\)\(^-\)\(^13\) Although the formation of grain macrostructures such as equiaxed and columnar grains can be simulated by both of these methods, the CA method has been used more frequently than the MC method because the MC method lacks a physical basis such as growth kinetics or preferential growth direction of a dendrite tip. In the CA method for simulating grain structure formation during solidification,\(^9\) the growth kinetics of a dendrite tip based on the KGT model.\(^14\) is introduced. However, the KGT model was originally developed for binary alloys, and the conventional CA method can not be used for simulation of the formation of macrostructures of multi-component alloy systems. Therefore, in our previous work, the PF-CA model was developed by coupling the phase-field (PF) method with the CA method for simulation of macrostructure formation in a casting such as equiaxed and columnar grains will be difficult.

Heterogeneous nucleation rates in solidifying aluminum-base multi-component alloys (AC2A, AC2B and AC8C commercial alloys) were determined as the form of the nucleation parameter. Unidirectional solidification experiments were carried out using the multi-component alloys and the nucleation parameters were determined by simulating the macrostructure similar to the experimentally observed one with a PF-CA model, which is a model developed by coupling a cellular automaton (CA) method with a phase-field (PF) method. A conventional casting experiment for the multi-component alloys was carried out, and prediction of the macrostructure of the castings with the PF-CA model has been carried out using the determined nucleation parameters from the unidirectional solidification. In the case of the AC2A commercial alloy, the simulated macrostructure was in good agreement with the experimentally observed one. In the case of the AC2B commercial alloy, although the simulated macrostructure near the mold wall was different from the experimentally observed one, the size of inner equiaxed crystals was similar to that observed in the experiment. In the case of the AC8C commercial alloy, the simulated macrostructure was not similar to the experimentally observed one. The disagreement between the results of the experiment and simulation might be due to a multi-phase solidification manner in the AC8C alloy.

KEY WORDS: aluminum-base multi-component alloy; heterogeneous nucleation; solidification; casting; phase-field method; cellular automaton method.
undercooling, standard deviation and maximum density of nuclei, for both heterogeneous nucleation phenomena in bulk liquid and at the mold wall. In this model, the physical meaning of each parameter is not clear because a complex relationship among their fitting parameters might exist. Therefore, a simple nucleation model is needed to predict the macrostructure of castings. In our pervious study\textsuperscript{15,16)} the following simple probability function for nucleation in bulk liquid as a function of undercooling, $\Delta T$, was proposed to simulate macrostructure formation in unidirectionally solidified Al–Si binary alloy casting:

$$ P = \left( \frac{\Delta T}{a} \right)^n $$

(1)

where $P$ is the probability of nucleation, $a$ is the maximum undercooling for nucleation, and exponent $n$ is the “nucleation parameter” for heterogeneous nucleation in undercooled melt. In the simulation, all of the liquid cells in the calculation domain are randomly scanned, and if the cell is an undercooled state, a random number, $q$, is generated ($0 \leq q \leq 1$). The value of $p$ is calculated from Eq. (1) by substituting the degree of undercooling, $\Delta T$, of the cell, and the state of the relevant cell is changed from a liquid state to solid state if $q$ is smaller than $p$. In this nucleation model, if a constant value of ‘$a$’ is used, the simulated structure varies from fully columnar to fully equiaxed morphology by arbitrary selection of the value of ‘$n$’. Nishiguchi et al. carried out CA simulation for macrostructure formation of a unidirectionally solidified Al–Si binary alloy casting by using this nucleation model.\textsuperscript{13)} The nucleation parameter, $n$, was determined by reproducing a macrostructure by CA simulation similar to the experimentally observed macrostructure.

However, investigation of heterogeneous nucleation parameters for multi-component alloys has not been carried out yet because the KGT model is adopted in the growth kinetics model of the CA method as described above and the KGT model can not be applied to the growth kinetics of a dendrite tip of multi-component alloys.

The purpose of the present work was to determine heterogeneous nucleation parameters for aluminum-base multi-component alloys and to carry out simulation of macrostructure formation in castings of aluminum-base multi-component alloys using the determined nucleation parameters. The validity of the use of a constant value of $a$ in Eq. (1) was also investigated.

2. **PF-CA Model**

The simulation procedure of the CA method used in the PF-CA model is basically the same as that of the CA technique developed by Rappaz and Gandin,\textsuperscript{9,10)} except for the nucleation model and the growth kinetics. Figure 1 shows a flowchart of the simulation using the present PF-CA model. The model is described in detail in below.

2.1. **Nucleation Model**

In the present model, it was assumed that the heterogeneous nucleation rate in the undercooled bulk liquid is proportional to the degree of undercooling, $\Delta T$, with a form of power function as shown in Eq. (1). Equation (1) expresses the probability of nucleation in each liquid CA cell and the use of Eq. (1) with probability form is convenient for carrying out the simulation. The real heterogeneous nucleation rate, $R_{Nu}$ (m$^{-2}$ s$^{-1}$), is expressed as follows:

$$ R_{Nu}(\Delta T) = \frac{1}{\Delta t \cdot V_{CA}} \left( \frac{\Delta T}{a} \right)^n $$

(2)

where $\Delta t$ is the time step and $V_{CA}$ is the area of a CA cell. This equation is a modification of Eq. (1) to give a physical meaning to the probability $P$. The appropriate value of ‘$n$’ was determined by reproducing the same macrostructure as those experimentally observed.

Equation (1) or (2) expresses the heterogeneous nucleation rate in the bulk liquid. On the other hand, at the mold wall, it is assumed that nucleation always occurs in the liquid CA cell in contact with the mold wall if the cell is in an undercooled state. This assumption is reasonable because it is expected that the potential of heterogeneous nucleation at the mold wall is considerably higher than that in the bulk liquid. In the present nucleation model, only two parameters, $i.e.$, $n$ and $a$, are required. Furthermore, an arbitrary value can be used for the parameter $a$ as discussed later. This nucleation model has a great advantage in comparison with the use of two Gaussian distributions in bulk liquid and at the mold wall because there is one nucleation parameter to be fitted.

2.2. **Growth Kinetics**

In the PF-CA model, the phase-field method was used for calculating growth kinetics of the dendrite tip of multi-component alloys instead of the KGT model. The phase-field method for a multi-component alloy with the thin interface limit parameter\textsuperscript{17)} was used in the present study. The phase-field equation and the solute diffusion equation for 2-dimensional calculation are as follows:

$$ \frac{\partial \phi}{\partial t} = M \left( \nabla^2 \phi - \frac{\partial f}{\partial \phi} \right) $$

(3)
\[
\frac{\partial c_i}{\partial t} = \nabla \left( \frac{D_i(\phi)}{f_{c_i}} \nabla f_i \right) \quad \ldots \quad (4)
\]

\[\sum_i c_i = 1 \quad \ldots \quad (5)\]

where \(\phi\) is the phase-field, \(M\) and \(\varepsilon\) are phase-field parameters, and \(c_i\) and \(D_i(\phi)\) are solute concentration and diffusion coefficient for the \(i\) alloy component, respectively. The free energy density of a solid–liquid mixture \(f(\phi, c_i)\) is expressed as follows:

\[
f(\phi, c_i) = h(\phi) f^3(c_i) + (1 - h(\phi)) f^4(c_i) + W g(\phi) \quad \ldots \quad (6)
\]

where

\[
h(\phi) = \phi^4(6\phi^2 - 15\phi + 10) \quad \ldots \quad (7)
\]

\[
g(\phi) = \phi^4(1 - \phi) \frac{1}{2} \quad \ldots \quad (8)
\]

In the above equations, \(f^3(c_i)\) and \(f^4(c_i)\) are the free energy density of solid and liquid phases, respectively. \(W\) is also a phase-field parameter.

The growth kinetics, i.e., undercooling, \(\Delta T\), vs. growth velocity, \(V\), relationship in multi-component alloys was determined from a 2-dimensional PF simulation of isothermal dendrite growth. The simulation was continued until a steady-state growth condition held, and then the growth velocity of a dendrite tip was measured for the case of several undercooling. This simulation condition corresponds to the KGT model with no temperature gradient \((G = 0)\). The calculated relationships between the undercooling, \(\Delta T\), and growth velocity of a dendrite tip, \(V\), is expressed by a function as given in Eq. (9).

\[
V(\Delta T) = \sum_m b_m (\Delta T)^m \quad (m = 1,2,\cdots,N) \quad \ldots \quad (9)
\]

where \(b_m\) and \(m\) are constants and given in Table 1.

2.3. Heat Transfer Calculation in Multi-component Alloy

In order to simulate the temperature field in a solidifying alloy casting, thermal properties of the alloy, such as thermal conductivity, density, specific heat, latent heat per unit mass, liquidus and solidus temperatures, and change in solid fraction, must be determined. In a binary alloy, these properties can be obtained from the published phase diagram. However, in a multi-component alloy, the determination of these properties is not easy. In this work, therefore, the enthalpy method was used as the method for heat transfer calculation for multi-component alloys because these features and properties of an alloy during solidification are involved in the temperature-enthalpy relationship of the alloy.

In the present work, the temperature-enthalpy relationships of multi-component alloys were calculated by using Thermo-Calc software. The thermo-dynamic database “ALDATA” developed by Thermotech was used in the calculation. In the enthalpy method, the temperature of each grid was determined from the change in enthalpy for each time step by using the calculated temperature-enthalpy relationship \(19,20\).

3. Experiment

Aluminum-base multi-component alloys; AC2A, AC2B and AC8C commercial alloys, were used in both the unidirectional solidification and conventional casting experiments. Nominal compositions of the commercial alloys used in the experiments are shown in Table 2. Figure 2 shows schematic illustrations of the experimental apparatuses. Figure 2(a) shows a mold made of an adiabatic material and a water-cooled box used for the unidirectional solidification experiment, and Fig. 2(b) shows a steel mold used for the conventional casting. Sample alloys were melted in an electric furnace and poured into the mold with superheat of 150 K. Change in the temperature of a solidified ingot for each experiment was measured by thermocouples.
set at the positions shown in Fig. 2.

In the solidification process of a casting, there are several mechanisms by which crystals multiply, and these mechanisms make it difficult to accurately predict solidified macrostructures. In the present work, only heterogeneous nucleation in an undercooled melt was considered. In the unidirectional solidification experiment, the mold was preheated up to 973 K in order to prevent nucleation on the mold wall, and the top of the mold was covered with a heated adiabatic cap just after the pouring procedure to prevent nucleation at the melt surface. In the conventional casting, adiabatic bricks on the top and bottom were preheated up to 973 K. The solidified ingots obtained in the unidirectional solidification were cut longitudinally at the center, and the solidified ingots obtained in the conventional casting were cut parallel to the thermocouples. These samples were polished and then etched with an acid reagent. The macrostructures of the ingots were metallographically examined, and the location of columnar to equiaxed transition (CET) was determined.

4. Calculation Procedure

2-Dimensional CA simulations of grain macrostructure formation were carried out. Figure 3 shows schematic illustrations of the simulation conditions. A network of square CA cells was laid on the cross section of an ingot, and a heat transfer grid was superimposed on the CA cells. One heat transfer grid contains 25 CA cells. The sizes of a heat transfer grid and a CA cell were 1.0×1.0 mm and 0.2×0.2 mm, respectively. For the heat transfer calculation, Eq. (10) was numerically solved for the unidirectional solidification and Eq. (11) was numerically solved for the conventional casting to calculate the change in enthalpy:

\[
\frac{\partial H}{\partial t} = \frac{k}{\rho} \left( \frac{\partial^2 T}{\partial r^2} + \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial z^2} \right) \quad \text{.........(10)}
\]

\[
\frac{\partial H}{\partial t} = \frac{k}{\rho} \left( \frac{\partial^2 T}{\partial x^2} + \frac{\partial^2 T}{\partial y^2} \right) \quad \text{.........(11)}
\]

where \( H \) is enthalpy per unit mass, \( k \) is thermal conductivity, \( \rho \) is density. The value of calculated enthalpy was converted to temperature by using the temperature-enthalpy curve. Table 3 shows material and thermophysical properties used in the simulation.21,22)

It should be noted that mass transfer due to diffusion or convection was not considered in the present PF-CA model. Hence, the definition of undercooling is the difference between the liquidus temperature for initial composition and the temperature of the relevant cell. The liquidus temperatures of the multi-component alloys were calculated by using the Thermo-Calc software.

In the simulation, the AC2A, AC2B and AC8C target al-

Table 3. Material and thermophysical properties used in the simulation.

<table>
<thead>
<tr>
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<th>Solid</th>
<th>Liquid</th>
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</thead>
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<tr>
<td>Thermal conductivity, ( \kappa ) (J/(m·s·K))</td>
<td>238</td>
<td>121</td>
</tr>
<tr>
<td>Density of Al, ( \rho_{Al} ) (Mg/m³)</td>
<td>2.70</td>
<td>2.39</td>
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<tr>
<td>Density of Si, ( \rho_{Si} ) (Mg/m³)</td>
<td>2.33</td>
<td>2.51</td>
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<tr>
<td>Density of Cu, ( \rho_{Cu} ) (Mg/m³)</td>
<td>8.93</td>
<td>7.93</td>
</tr>
<tr>
<td>Density of Mg, ( \rho_{Mg} ) (Mg/m³)</td>
<td>1.74</td>
<td>1.59</td>
</tr>
<tr>
<td>Heat transfer coefficient, ( \kappa ) (W/(m²·K))</td>
<td>41.8</td>
<td></td>
</tr>
<tr>
<td>Ingot-Air, ( h_{\text{air}} ) (t : time)</td>
<td>5.67×10⁹</td>
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</tr>
<tr>
<td>Ingot-Chill, ( h_{\text{chill}} ) (t : time)</td>
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< Unidirectional solidification >

<table>
<thead>
<tr>
<th></th>
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<th>AC2B</th>
<th>AC8C</th>
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</thead>
<tbody>
<tr>
<td>( h_{\text{air}} )</td>
<td>5.0×10⁻⁵</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( h_{\text{chill}} )</td>
<td>2.5×10⁻⁵</td>
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< Conventional casting >

<table>
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<tr>
<th></th>
<th>AC2A</th>
<th>AC2B</th>
<th>AC8C</th>
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<tbody>
<tr>
<td>( h_{\text{air}} )</td>
<td>5.0×10⁻⁵</td>
<td></td>
<td></td>
</tr>
<tr>
<td>( h_{\text{chill}} )</td>
<td>2.5×10⁻⁵</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

Stefan-Bolzmann coefficient, \( \sigma \) (W/(m²·K⁴)) 5.67×10⁻⁸
Interfacial energy, \( \sigma \) (J/mol) 0.093
Molar volume, \( V_m \) (m³/mol) 7.7×10⁻⁶
Diffusion coefficient for Al in solid, \( D_{Al} \) (m²/s) 3.0×10⁻¹⁰
Diffusion coefficient for Al in liquid, \( D_{Al} \) (m²/s) 6.1×10⁻⁹
5. Results and Discussion

5.1. Estimation of Heterogeneous Nucleation Parameters

Figure 4 shows the cooling curves during unidirectional solidification of the AC2A commercial alloy for (a) the experiment and (b) simulation. Dotted lines in Fig. 4 show liquidus temperature, \( T_L = 890 \) K. Simulated cooling curves near the liquidus temperature are in good agreement with those obtained by the experiment. This is important because the CET occurs at the position of the solidification front, and the present PF-CA model could accurately simulate the solidification front.

Figure 5 shows experimentally observed and simulated macrostructures in the aluminum-base commercial alloy ingots. In the experiment, feather crystals grew from the bottom of the macrostructures of AC2A and AC8C alloys. It has been reported that feather crystals sometimes form in a unidirectionally solidified ingot when the pouring temperature is relatively high.\(^{23,24}\) In the present work, it was assumed that the region of feather crystals corresponds to the region of columnar grains, and the transition from feather crystals to equiaxed grains was regarded as the CET. In the simulated macrostructures, the CET occurred in the upper region of the ingots as observed in the real structures obtained by the experiment. The values of nucleation parameter \( n' \) were determined for each alloy by simulating the positions of the CET in the macrostructure observed in the experiment. When the value of \( \alpha \) was constant (\( \alpha = 40 \)), the values of \( n' \) were 6.4 (AC2A), 6.5 (AC2B) and 5.5 (AC8C).

Table 4 shows the combinations of the values of \( \alpha \) and \( n' \) used in the present simulation. In order to simulate macrostructures similar to the experimental ones, the values of \( \alpha \) and \( n' \) must be determined simultaneously.

Figure 6 shows the simulated macrostructures of the AC2A commercial alloy by using different combinations of values of \( n' \) and \( \alpha \). It was found that the position of CET can be simulated with different combinations of values of \( n' \) and \( \alpha \). Figure 7 shows the relationship between undercooling and nucleation rate for the three combinations of values of \( n' \) and \( \alpha \) used in the simulation shown in Fig. 6. Figure 7(a) shows all range of undercooling of 40 K, and Fig. 7(b) is a magnified view at the low undercooling region of Fig. 7(a). For all range of undercooling in Fig. 7(a), it seems that the shapes of three curves are different. However, the shapes of the three curves are almost the same in the low undercooling region shown in Fig. 7(b). Figure 8 shows the undercooling distribution when heterogeneous nucleation occurred in the simulation of unidirectional solidification of an AC2A commercial alloy ingot. The combination of nucleation parameters is \( \alpha = 40 \) and \( n' = 6.4 \). It can be seen that most nuclei are formed with relatively small undercooling of about 5 K except for the region near the chill block. It
seems that the difference in the shape of the undercooling vs. nucleation rate relationship in the high undercooling region in Fig. 7(a) is not problem to simulate the macrostructure of a casting. These results demonstrate that the use of the combination of a fixed value of ‘a’ and a variable ‘n’ is effective for the simulation of solidification structure formation.

5.2. Prediction of Macrostructures for Conventional Casting

Figure 9 shows experimentally observed and simulated macrostructures of aluminum-base commercial alloy ingots for conventional casting, respectively. In the simulation of conventional casting, a combination of nucleation parameters, determined by the simulation of unidirectional solidification, were used (see Table 4). The position of CET in the simulated macrostructure of the AC2A commercial alloy (Fig. 9(1)) was in good agreement with the experimentally observed one (Fig. 9(1)(a)), and simulated grain size
was also similar to the experimental ones. In this case, these results demonstrate that the nucleation parameter determined from a unidirectional solidification experiment is effective for the prediction of macrostructure of conventional casting.

In the macrostructures of the AC2B commercial alloy (Fig. 9(2)), the experimentally observed macrostructure (Fig. 9(2)(a)) was almost equiaxed crystals, and the grain size near the mold was small. In the simulation (Fig. 9(2)(b)), columnar grains grew from the mold wall in the region near the mold wall. However, the size of inner equiaxed crystals was similar to that of the experimental ones. In the experimentally observed macrostructure of AC8C commercial alloy (Fig. 9(3)(a)), the etched structure was not so clear, but it could be seen that the structure was composed of mostly columnar grains and some feather crystals. The region of equiaxed crystals was identified as the square area with a white dotted line in Fig. 9(3)(a). In this case, the simulated position of CET did not agree with the experimental result (Fig. 9(3)(b)).

In the case of the solidification process of the AC8C alloy, the amount of secondary and ternary phases that were formed in the spaces between primary α-dendrites is large in comparison with that of the AC2A and AC2B alloys. The disagreement between the simulated and experimentally observed macrostructures of the AC8C alloy might be due to the fact that the conventional CA method used in the present simulation can not handle the process of multiphase solidification. In the conventional CA method, only the growth process of the primary phase can be simulated and it is impossible to simulate the complex multiphase solidification process of multi-component alloys.

6. Conclusions

Heterogeneous nucleation parameters of aluminum-base multi-component alloys (AC2A, AC2B and AC8C alloys) were determined from a unidirectional solidification experiment and simulation of macrostructure formation using a PF-CA model. A simulation of the macrostructure formation of multi-component alloys during solidification for the conventional casting process was carried out using solidification parameters determined from the experiment and simulation of unidirectional solidification. In the case of the AC2A commercial alloy, the simulated macrostructure was in good agreement with the experimental one. In the case of the AC2B commercial alloy, although the experimentally observed structure near the mold wall was not in agreement with the simulated one, the size of the equiaxed crystals located in the inner region of the ingot was similar to the simulated macrostructure. In the case of the AC8C commercial alloy, the simulated macrostructure was not similar to the experimentally observed structure. The CA method used in the present simulation might not be able to handle the multiphase solidification of a multi-component alloy such as AC8C alloy. However, it can be concluded that the present PF-CA model has potential for predicting the macrostructure of a multi-component alloy casting.

REFERENCES