Three-dimensional Cellular Automaton Model for the Prediction of Microsegregation in Solidification Grain Structures

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A 3-D cellular automaton finite difference (3-D CAFD) model to predict the solidification grain structure and the microsegregation was developed. In the present model, a new approach to calculate the solute concentration distribution was developed. Moreover, the CA cell including the grain boundary was newly defined. The probabilistic approach was adopted as the nucleation model, and the decentered octahedron growth algorithm was adopted as the grain growth model. The growth kinetics of the dendrite tip to calculate the growth of the dendrite envelope was calculated by a 3-D Kurz-Giovanola-Trivedi (KGT) model. In the 3-D KGT model, the local undercooling estimated by the local temperature and local solute concentration was used. To evaluate the validity of the present model, we carried out simulations of the solidification grain structure under directional solidification. The different solidification grain structures were obtained by the different solute concentration distributions. In addition, we compared our model with the microsegregation predicted by Scheil's equation. The simulated results of microsegregation by the present model were in fairly good agreement with those by Scheil's equation. From these results, we confirmed that the calculation of solute concentrations has to be considered and that the present model can simultaneously simulate microstructure and microsegregation.

KEY WORDS: cellular automaton method; microsegregation; grain structure; simulation; solidification.

1. Introduction

The Cellular Automaton (CA) method is an efficient tool to predict solidification grain structures. Gandin *et al.* have been developing a mesoscopic-scale (over *mm* scale) CA model to simulate solidification grain structures since the early 1990 s.^{1–5)} First, a two dimensional CA (2-D CA) model¹⁾ was proposed for the modeling of dendritic grain structures, and then it was coupled with a finite element (FE) heat flow calculation, which is called the 2-D CAFE model.^{2,3)} After that, a three dimensional CAFE (3-D CAFE) model was developed.^{4,5)} When the CA model is coupled with a finite difference (FD) heat flow calculation, the model is called the CAFD model. It is well known that the CAFE and CAFD models have been shown to predict grain structures successfully in various solidification processes.^{6–9)} Recently, microscopic-scale (less than *mm* scale) CA models have been developed by some researchers,^{10–17)} which are often called modified CA model^{10,11)} or front tracking model.^{12,14–16)} This microscopic-scale CA model can simulate the dendritic morphologies like a phase-field (PF) method^{18–22)} and have been proposed in 2-D and 3-D.^{11,15,16)} However, predicting the dendritic grain structures for solidification of large samples such as ingots, billets, and slabs will be difficult because of the high computational costs, especially in 3-D. Therefore, the mesoscopic-scale CA model

will be the most practical and efficient tool to predict the solidification grain structures.

The mesoscopic-scale CA model consists of the nucleation model and the grain growth model^{1–5)} and is coupled with FE or FD heat flow calculations. The nucleation model is based on the instantaneous or continuous nucleation theories. The grain growth model is based on the theory of the growth kinetics of dendrite such as the KGT model²³⁾ and the LKT model.²⁴⁾ In the CA model, a domain is divided into fine CA cells, and the probability of nucleation and the growth velocity of grains are essentially calculated by the local undercooling of a CA cell. The local undercooling is estimated as the difference of liquidus temperature and local temperature of a cell. The local temperatures are obtained by the FE or FD heat flow calculations. In the conventional model,^{1–5)} the liquidus temperature is given at the initial setting of the calculation: it is just the liquidus temperature at the initial composition of the calculation. However, the liquidus temperature will differ as a function of the solute concentration of each CA cell. Thus, the liquidus temperature has to be determined by the solute concentration of the CA cell. For example, in an alloy for which the equilibrium partition coefficient is less than one, the solute is rejected at the solid-liquid interface during solidification. Thus, the solute concentration in the liquid becomes higher and that in the solid becomes lower as it is solidified. Therefore, the liquidus temperature has to be lower for the liquid region having the higher solute concentration: this corresponds to smaller undercooling in the liquid region having the higher solute

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concentration. Nucleation and growth can be simulated correctly by using a local undercooling estimated from temperature and solute concentration. Therefore, to quantitatively simulate the solidification grain structures, the solute concentration distribution that is not considered in the conventional model has to be calculated in the model. In such a model, it is possible to simultaneously predict solidification grain structures and the microsegregation pattern by calculating the solute concentration in the domain.

One of the aims of this work was to present a new approach of a 3-D CAFD model that could simulate grain structures, temperature and concentration fields. In the present model, we adopted the nucleation model using the probabilistic approach proposed by Rappaz and Gandin,¹⁾ and the growth model using the decentered octahedron growth algorithm proposed by Gandin and Rappaz.⁴⁾ In addition, the 3-D KGT model²³⁾ was used as the growth kinetics of dendrite tip. To simulate microsegregation, we developed a new approach to estimate the solute concentration distribution in grain structures. Therefore, in the calculation of nucleation and growth, the undercooling estimated by local temperature and local concentration for each cell was used. From the simulated results in this model, we investigated the potential to predict the microsegregation pattern using the present model.

2. Model

In our 3-D CAFD model, the CA model for nucleation and grain growth was coupled with FD model for heat flow. The heat flow was calculated at the level of a coarse FD grid, whereas nucleation and grain growth were calculated at the fine CA cell level. In addition, the solute concentration was also calculated at the fine CA cell level.

2.1. Heat Flow Calculation

The 3-D heat flow was calculated at the macro level using a cubic FD grid, and the enthalpy method^{2,5)} was adopted to calculate the heat flow, considered the latent heat during solidification. The local temperatures at the CA cell q were deduced from temperatures of the eight nearest nodes obtained by the FD heat flow calculation. **Figure 1** shows a schematic illustration of the relationship between the local temperature of CA cell q and the temperatures at the nearest nodes of FD grids. The local temperatures of CA cell q , T_q^t , were estimated from the temperatures at the nearest nodes of FD grids by linear interpolation as follows:

$$T_q^t = \sum_{i=1}^8 \phi_i T_i^t \quad \dots\dots\dots (1)$$

where t is a given time, ϕ_i is the interpolation coefficient, T_i^t is the temperature at the nearest nodes of FD grids. In the enthalpy method, the variations of enthalpy at the FD node locations within each time step, ΔH , were calculated using the heat flow equation, and the new enthalpy, $H^{t+\Delta t}$, was calculated by $H^t + \Delta H$. After that, the new temperatures at the FD node locations, $T^{t+\Delta t}$, were determined using the relationship between enthalpy and temperature. In addition, the new solid fraction at the FD node locations, $f_s^{t+\Delta t}$, were also determined using the relationship between temperature and solid fraction. These relationships were obtained from ther-

Fig. 1. Schematic illustration showing the relationship between the local temperature of CA cell and the temperatures at the nearest nodes of FD grids.

modynamic data. In this model, the relationship among enthalpy, temperature and solid fraction was calculated using PANDAT software. In the simulations, this relationship calculated at the initial composition was used.

2.2. Nucleation Model

The nucleation model adopted was the probabilistic approach proposed by Rappaz and Gandin.¹⁾ This model is based on an instantaneous nucleation and Gaussian distribution of nucleation sites and is assumed to model the grain density increase, dn , that is induced by an increase in the undercooling, $d(\Delta T)$. The distribution, $dn/d(\Delta T)$, is described as follows:

$$\frac{dn}{d(\Delta T)} = \frac{n_{max}}{\sqrt{2\pi}\Delta T_{\sigma}} \exp \left[-\frac{1}{2} \left(\frac{\Delta T - \Delta T_{max}}{\Delta T_{\sigma}} \right)^2 \right] \quad \dots\dots (2)$$

where n_{max} is the maximum nucleation density, ΔT_{max} is the mean undercooling, and ΔT_{σ} is the standard deviation. The different distributions are often used to describe heterogeneous nucleation at the mold (or cooling chill) surface and in the bulk of the melt. The maximum nucleation density at the mold surface, n_{max}^m , has the units of m^{-2} , whereas that in the bulk of the melt, n_{max}^b , has the units of m^{-3} . The maximum undercooling and the standard deviation were also determined both at the mold surface and in the bulk of the melt: ΔT_{max}^m , ΔT_{max}^b , ΔT_{σ}^m , and ΔT_{σ}^b . To quantitatively simulate the solidification grain structures, these parameters have to be determined experimentally for a given alloy.

2.3. Grain Growth

The 3-D CA growth algorithm has to be able to reproduce the preferential $\langle 100 \rangle$ growth directions of fcc dendrites and their growth kinetics. The 3-D CA growth algorithm adopted was the decentered octahedron growth algorithm proposed by Gandin and Rappaz.⁴⁾ This growth algorithm was based on the growth of an octahedron bounded by (111) faces and assumed that the vertex of an octahedron corresponded to six dendrite tips. The same growth kinetics was used at the six dendrite tips of a given octahedron. Once the growing octahedron captured the cell center of one of its neighbors, the state index of the captured cell was then

Fig. 2. Relationship among dendrite tip growth velocity, temperature and composition of Al-Si binary alloy obtained by the 3-D KGT model.

switched to that of the parent cell. The growth of a new octahedron was considered later, and the center position of a new octahedron was different from that of the old octahedron: it was a “decentered octahedron.” Details of this algorithm can be found in Ref. 4). It is noted that the growth of an octahedron, that is, the evolution of dendrite envelope, depended on the growth velocity of dendrite tips. The growth velocity of a dendrite tip was calculated by the 3-D KGT model, and it was essentially given as a function of the local undercooling of the CA cell. In the present model, the growth velocity, $v(\Delta T)$, was not given as a function of the local undercooling only. It was given as a function of the local temperature and the local solute concentration: $v(T, c)$ was used. Most researchers use a simplified form (using either a polynomial law or power law) of the above growth velocity by a direct interpolation of the relationship between the growth velocity and the local undercooling. However, in the present model, we did not use such a simplified form. **Figure 2** shows the relationship among the growth velocity, temperature, and composition for a hypoeutectic Al-Si binary alloy calculated using the 3-D KGT model. The range of composition was from 1.65 to 12.6 wt% Si. It is noted that the higher growth velocity could be obtained when temperature and composition were lower. In the present model, before the simulation for a given alloy, the v - T - c relationship was prepared in advance. In the CA simulations, this relationship was used as a database for a given alloy.

2.4. Grain Boundary

In the present model, five states of the CA cell which were liquid, solid, active mushy, inactive mushy, and grain boundary, were used to distinguish between the concentration in grains and that out of grains. **Figure 3** shows a schematic illustration to explain the five states of the CA cell. Liquid cells are the cells where only liquid phase existed, solid cells are the cells where the primary phase coexisted with the eutectic phase, and mushy cells are the cells where the primary phase coexisted with the liquid phase. Moreover, the state of mushy cells is distinguished as “active” or “inactive.” “Active” means mushy cells included in the growing dendrite envelope, and “inactive” means the mushy cells fully surrounded by “active” or “inactive” mushy cells.

Fig. 3. Schematic illustration showing five states of CA cell defined in the present model.

Grain boundary cells are the cells that formed the grain boundaries after the cells became fully solid state. Thus, the thickness of a grain boundary is not equal to the size of CA cell. Grain boundary cells will be given between the inactive mushy cells. The transition to grain boundary cells was carried out as follows: First, the active mushy cell of a grain came next to those of other grains due to grain growth. After that, the “active” mushy cells became “inactive” by the change in the state of the surrounding cells. Finally, one of the “inactive” mushy cells changed into a grain boundary cell.

By considering the grain boundary cell in the model, the solute concentrations in grains and out of grains could be different solute concentrations after solidification. This concept was important for simulating the microsegregation in the grain size level.

2.5. Solute Concentration

The solute concentration was calculated by assuming that there was no diffusion in the solid phase and complete mixing in the liquid phase, as in the Scheil-type relationship. Otherwise, the assumption of partial mixing in the liquid phase was strictly used in the model. The diffusion equation was not solved directly for solid or for liquid. **Figure 4** shows a schematic illustration of the model used to calculate the solute concentration for the present research. In the model, two states of solid and liquid were defined to calculate the solute concentration. Solid and liquid states were distinguished by the critical solid fraction, f_{sc} . Thus, the mushy region was included in both solid and liquid states. The mushy region where the solid fraction was smaller than f_{sc} corresponded to the liquid state, whereas the mushy region

Fig. 4. Schematic illustration of the model used for calculating the solute concentration in the present model.

where the solid fraction was larger than f_{sc} corresponded to the solid state. The solute concentration of the liquid state, C_L , was homogeneous for the whole CA cells of liquid state. This is because the solute in liquid was assumed to be mixed by convection. The solute concentration of the solid state was determined for the active mushy cell as follows: The active mushy cells of the solid state coexisted with solid and liquid phases. The solute concentration of these cells was calculated by the average concentration in the mixed solid and liquid, C_{mix} . C_{mix} was defined as follows:

$$C_{mix} = (1 - f_{sc})C_L + k_{eff}f_{sc}C_L \quad \dots\dots\dots (3)$$

where k_{eff} is the effective partition coefficient.²⁵⁾ k_{eff} was defined as follows:

$$k_{eff} = \frac{k_0}{k_0 + (1 - k_0)\exp(-V\delta / D_L)} \\ = \frac{k_0}{k_0 + (1 - k_0)\exp(-2\delta / \delta_L)} \quad \dots\dots\dots (4)$$

where k_0 is the equilibrium partition coefficient, V is the growth velocity of the solidification front, D_L is the diffusion coefficient in liquid, δ_L is the diffusion layer thickness, and δ is the diffusion layer thickness for the partial mixing. We used the assumption of $\delta = \delta_L/2$ in this simulation: the solute concentration of the region over δ was homogeneous by convection.

In the calculation, when the solid fraction of a CA cell was over the value of f_{sc} , the concentration of the CA cell became C_{mix} calculated by Eq. (3): the cell was regarded as the mushy cell in the solid state shown in Fig. 4. After the transition of CA cells over the value of f_{sc} finished, the C_L was calculated. The value of C_L was determined by mass balance in the calculation domain, and was homogeneous for all of CA cells in liquid state shown in Fig. 4. In case of $k_0 < 1$, the value of C_L became gradually higher during solidification, and it was highest at the end of solidification. The solute concentration distribution formed during solidification affected the formation of solidification grain structures.

Fig. 5. Schematic illustration of the calculation domain for 3-D directional solidification simulations.

In the present model, it is noted that the f_{sc} was also one of parameters used to quantitatively simulate the solute concentration distribution.

3. Results and Discussion

Figure 5 shows a schematic illustration of the calculation conditions of a directional solidification. The calculations were performed on a rectangular parallelepiped domain of $100 \times 100 \times 200$ mesh with uniform CA cell size $\Delta x_{CA} = 100 \mu\text{m}$ and that of $20 \times 20 \times 40$ mesh with uniform FD mesh size $\Delta x_{FD} = 500 \mu\text{m}$. The initial temperature in the entire domain was assumed to be homogeneous with a constant temperature, T_{ini} . The cooling chill was assumed to be set on the bottom of the calculation domain. The temperature of the cooling chill was always constant, T_{chill} , during calculations. Zero-flux boundary conditions were used for temperature and solute concentration on four side and top surfaces.

First, we discuss the simulation results for solidification grain structures. Figure 6 shows the final solidification grain structures of Al-3wt%Si binary alloy calculated using the present 3-D CAFD model and various values of f_{sc} . The materials properties and the calculation parameters used in this simulation are shown in Table 1. The solidification grain structures shown in Figs. 6(a), 6(b), and 6(c) were for the values of f_{sc} of 0, 0.2, and 0.4, respectively. The nucleation parameters were the same for all calculations. The solidification grain structure of $f_{sc} = 0$ (Fig. 6(a)) corresponded to the one calculated by the conventional method where the calculation of the solute concentration was not considered: the solute concentrations were always the initial composition in the whole cells. In the solidification grain structure of $f_{sc} = 0$, a structure with only columnar grains was obtained, while, in those of $f_{sc} = 0.2$, and 0.4, structures with columnar and equiaxed grains were obtained. The formation of structures was affected by the rate of nucleation and grain growth. If grain growth was preferred over the rate of nucleation for a region of an undercooling melt, the coarse equiaxed or the columnar structures was formed.

Fig. 6. Final solidification grain structures of Al-3wt%Si binary alloy calculated using the present model and various values of f_{sc} : (a) $f_{sc} = 0.0$, (b) $f_{sc} = 0.2$, and (c) $f_{sc} = 0.4$.

Table 1. Material properties and calculation parameters used in our simulations.

Initial composition, C_0 [wt.%Si]	3.0
Initial temperature of melt, T_{ini} [°C]	645
Chill temperature, T_{chill} [°C]	50
Thermal conductivity of solid, κ_s [W/Km]	238
Thermal conductivity of liquid, κ_l [W/Km]	95
Density of solid, ρ_s [g/cm ³]	2.70
Density of liquid, ρ_l [g/cm ³]	2.39
Equilibrium partition coeff., k_0	0.12
Heat transfer coeff. (chill-melt), h [W/Km ²]	1 000
CA cell size, Δx_{CA} [m]	1×10^{-4}
FD grid size, Δx_{FD} [m]	5×10^{-4}
Critical solid fraction, f_{sc}	0, 0.2, 0.3, 0.4
Nucleation parameter in the bulk of the melt	
Standard deviation, ΔT_σ [K]	0.5
Mean undercooling, ΔT_{max} [K]	7.0
Maximum nucleation density, n_{max} [m ³]	1×10^{-5}
Nucleation parameter at the chill surface	
Standard deviation, ΔT_σ [K]	0.1
Mean undercooling, ΔT_{max} [K]	1.0
Maximum nucleation density, n_{max} [m ²]	1×10^{-4}

Conversely, if the rate of nucleation is preferred to the growth of grains, fine equiaxed structures were formed. In case of $f_{sc} = 0$, the growth of grains was preferred to the rate of nucleation at the end of solidification, while, in cases of $f_{sc} = 0.2$ and 0.4 , the rate of nucleation was preferred to the growth of grains. In $f_{sc} = 0.2$ and 0.4 , the solute enriched around the top of the domain at the end of solidification, so the grain growth velocity was lower than that at the beginning of solidification. As a result, the equiaxed grains formed in front of columnar grains. On the other hand, in $f_{sc} = 0$, the solute concentration was homogeneous in the domain, and thus, the growth velocity of grains remained high at the end of solidification. Therefore, the columnar grains continued to grow on the top of the domain. From these results, to

quantitatively simulate the solidification grain structures using a CA model, it was very important to consider the calculation of the solute concentration in the model.

Second, we discuss the simulation results for solute concentration distribution. **Figure 7** shows the average silicon concentration profiles for final solidification grain structure of $f_{sc} = 0.2$. All of cells were solid cell consisted of the primary and eutectic phases or grain boundary. Thus, the silicon concentration for each cell was the average value of the phases. The average silicon concentration of a solid cell, C_s , was calculated as follows:

$$C_s = C_{pri} f_{pri} + C_{eut} f_{eut} \quad (5)$$

where C_{pri} and C_{eut} are the silicon concentrations of primary and eutectic phases in a cell, respectively, and f_{pri} and f_{eut} are the fractions of primary and eutectic phases in a cell, respectively. The relationship between f_{pri} and f_{eut} was $f_{pri} + f_{eut} = 1$ in a cell. **Figure 7(a)** is the 3-D view of the average silicon concentration of the final solidification grain structure. The region of higher silicon concentration was observed at the top of domain. The structure of this region consisted of equiaxed grains. In the region of columnar grains, the silicon concentration of grains was clearly different from that of grain boundaries. **Figure 7(b)** is the average silicon concentrations and solidification grain structures of the cross sections for regions of equiaxed grains and of columnar grains. In the region of equiaxed grains, the regions of the various silicon concentrations were randomly observed, and it is noted that the regions of the highest silicon concentration (red regions) were the last to solidify. On the other hand, in the region of columnar grains, the silicon concentration in grains was clearly different from that of grain boundaries. The size of grains was almost homogeneous. From these results, we confirmed that the present model was able to simulate microsegregation patterns at a microstructure level.

Finally, we discuss the quantitative evaluation of microsegregation by the present model. **Figure 8** shows the relationships between the average silicon concentrations calculated using various values of f_{sc} and the distance from the bottom of the domain. For comparison, the profile of the silicon concentration estimated by using Scheil's equation²⁶⁾ is also shown in **Fig. 8**. We modified Scheil's equation to

Fig. 7. Average silicon concentration distributions for final solidification grain structure of $f_{sc} = 0.2$. (a) 3-D view and (b) cross sections for the regions of equiaxed and columnar grains. (Online version in color.)

Fig. 8. Relationship between average silicon concentration distribution and distance from the bottom of domain. (a) $f_{sc} = 0.2$, (b) $f_{sc} = 0.3$, and (c) $f_{sc} = 0.4$.

compare with our simulations. The silicon concentration mixed solid and liquid was calculated as follows:

$$C_s = k_m C_0 (1 - f_s)^{(k_m - 1)} \quad (6)$$

where k_m is the partition coefficient modified as the ratio of concentrations of mushy region, C_m^e , and liquid, C_L^e , at the equilibrium state.

$$k_m = \frac{C_m^e}{C_L^e} = \frac{f_{sm} C_s^e + (1 - f_{sm}) C_L^e}{C_L^e} = 1 - (1 - k_0) f_{sm} \quad (7)$$

where f_{sm} is the fraction of primary solid phase in the mushy region at equilibrium state. The values of k_m will be larger than that of k_0 when $f_{sm} < 1$ for the condition of $k_m < 1$. Figs. 8(a), 8(b), and 8(c) are the profiles of the average silicon concentration versus the distance from the domain bottom for $f_{sc} = 0.2$, 0.3, and 0.4, respectively. The C_s in the simulations was the average of the silicon concentrations of 100×100 cells at a distance from the domain bottom. At all values of f_{sc} , negative segregation was observed in the region about 0.014 m from the bottom of the domain, while positive segregation was observed in the region over 0.014 m. In the initial stage of solidification, the grain selection occurred by many equiaxed grains. Therefore, in this stage, since many fine grains are formed, the degree of negative segregation was small. The degree of negative and positive segregation increased with increasing f_{sc} : in the region of negative segregation, the silicon concentration in $f_{sc} = 0.2$ became lower than that in $f_{sc} = 0.3$ and 0.4, and in the region of positive segregation, the silicon concentration in $f_{sc} = 0.4$ became higher than that in $f_{sc} = 0.2$ and 0.3. Compared with the profiles of Scheil's equation, except for the region of the grain selection, the silicon concentration distributions simulated in the present model were in fairly good agreement with those calculated by Scheil's equation for all of the conditions of f_{sc} . This agreement was obtained when the values of f_{sm} of 0.1, 0.15, and 0.2 were used for the simulations of f_{sc} of 0.2, 0.3, and 0.4, respectively. As a result, the relationship between f_{sm} and f_{sc} was $f_{sm} = 0.5 f_{sc}$. No obvious theoretical explanation exists for the relationship of $f_{sm} = 0.5 f_{sc}$, howev-

er, we confirmed that the simulated results of microsegregation by the present model qualitatively agree Scheil's equation, and that the present model has the potential to quantitatively predict the solidification grain structures and microsegregations. Comparisons with experimental data are required to determine if the model can make a successful quantitative prediction of solidification grain structures and the microsegregations. In the present model, the most important parameter to evaluate will be f_{sc} . For the practical use of the present model, we must investigate the relationship between f_{sc} and the microsegregation pattern, and compare with experimental data.

4. Conclusions

A new approach using a 3-D CAFD model to predict solidification grain structure and microsegregation was presented. In the present model, a 3-D KGT model²³⁾ was used for the growth kinetics of a dendrite tip, and the undercooling by the local temperature and the local solute concentration at a CA cell was considered to estimate the growth kinetics of a dendrite tip. We developed a new approach to calculate the solute concentration. In the model to calculate the solute concentration in grains, a new parameter f_{sc} was defined. Moreover, the grain boundary cells were newly defined in the model.

We carried out simulations for directional solidification of an Al-3wt%Si binary alloy using the present model and discussed the validity of our model from the simulated results. The results of the validity discussion are as follows:

(1) To quantitatively simulate the solidification grain structures, the calculation of solute concentrations have to be considered in the model because the solute concentration distributions are affected in the dendrite growth kinetics.

(2) The simulated results of microsegregation by our new approach were in fairly good agreement with those by Scheil's equation. From this result, we confirmed the valid-

ity of the present model to quantitatively predict microsegregations at the solidification grain level. In addition, the present model can simultaneously simulate microstructure and microsegregation.

The 3-D CA model to simulate the grain structure was already developed by Gandin and Rappaz.⁴⁾ However, it is important to combine the model with a FD (or FE) aspect to predict microsegregation as was done in this study. Such a 3-D CAFD (or CAFE) model will be of considerable help for control of grain structure and segregation in complex casting products.

REFERENCES

- 1) M. Rappaz and Ch.-A. Gandin: *Acta Metall. Mater.*, **41** (1993), 345.
- 2) Ch.-A. Gandin and M. Rappaz: *Acta Metall. Mater.*, **42** (1994), 2233.
- 3) Ch.-A. Gandin and M. Rappaz: *Acta Metall. Mater.*, **44** (1996), 3339.
- 4) Ch.-A. Gandin and M. Rappaz: *Acta Metall. Mater.*, **45** (1997), 2187.
- 5) Ch.-A. Gandin, J.-L. Desbiolles, M. Rappaz and Ph. Thevoz: *Metall. Mater. Trans. A*, **30A** (1999), 3153.
- 6) Y. Natsume, N. Nishiguchi, K. Ohsasa and T. Narira: *J. JFS*, **77** (2005), 31.
- 7) Y. Natsume and K. Ohsasa: *ISIJ Int.*, **46** (2006), 896.
- 8) M. Yamazaki, Y. Natsume, H. Harada and K. Ohsasa: *ISIJ Int.*, **46** (2006), 903.
- 9) H. Ishida, Y. Natsume and K. Ohsasa: *ISIJ Int.*, **48** (2008), 1728.
- 10) M. F. Zhu and C. P. Hong: *ISIJ Int.*, **41** (2001), 436.
- 11) M. F. Zhu and C. P. Hong: *ISIJ Int.*, **42** (2002), 520.
- 12) L. Nastac: *Acta Mater.*, **47** (1999), 4253.
- 13) L. Beltran-Sanchez and D. M. Stefanescu: *Metall. Mater. Trans. A*, **34A** (2003), 367.
- 14) L. Beltran-Sanchez and D. M. Stefanescu: *Metall. Mater. Trans. A*, **35A** (2004), 2471.
- 15) M. F. Zhu and D. M. Stefanescu: *Acta Mater.*, **55** (2007), 1741.
- 16) S. Pan and M. F. Zhu: *Acta Mater.*, **58** (2010), 340.
- 17) W. Wang, P. D. Lee and M. McLean: *Acta Mater.*, **51** (2003), 2971.
- 18) R. Kobayashi: *Physica D*, **63** (1993), 410.
- 19) A. Karma and W. -J. Rappel: *Phys. Rev. E*, **57** (1998), 4323.
- 20) M. Ohno and K. Matsuura: *Phys. Rev. E*, **79** (2009), 031603.
- 21) Y. Natsume: *R&D Kobe Steel Eng. Rep.*, **58** (2008), 35.
- 22) Y. Natsume and H. Ishida: Proc. of ICAA12, JILM, Tokyo, (2010), 1746.
- 23) W. Kurz, B. Giovanola and R. Trivedi: *Acta Metall.*, **34** (1986), 823.
- 24) J. Lipton, W. Kurz and R. Trivedi: *Acta Metall.*, **35** (1987), 957.
- 25) J. A. Burton, R. C. Prim and W. P. Slichter: *J. Chem. Phys.*, **21** (1953), 1987.
- 26) E. Scheil: *Z. Metallkd.*, **34** (1942), 70.