Numerical simulations of solidification structures and macrosegregation by a cellular automaton model coupled with flow calculations

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Abstract. In this study, a numerical model was developed, and direct simulations were performed to predict solidification grain structures and macrosegregation based on a three-dimensional cellular automaton finite difference (CAFD) method coupled with flow calculation of natural convection and shrinkage flow. First, to evaluate the model coupled with natural convection, simulations of unidirectional solidification for Al-10wt% Mg alloy were performed. Mg-rich plumes rising in the melt were seen due to subsequent upward flow, and Mg-rich channels forming in the mushy zone were observed. Columnar grains were then formed, and they became coarse afterwards. Next, to evaluate the model coupled with shrinkage flow, simulations of casting Al-10wt% Cu alloy in a unique mold, which can form macrosegregation in the central region of the small ingot, were performed. The bridging of columnar grains formed during solidification, and the positive segregation was generated in the region below the bridging. Thus, the main factor for this macrosegregation is the shrinkage flow with bridging. From the comparison of simulation results with and without the chill for the unique mold, it was established that the shrinkage flow and the bridging of solidification structures play an important role in macrosegregation.

1. Introduction
The cellular automaton (CA) model is one of the tools used to efficiently predict solidification structures. This model coupled with a finite element (FE) heat flow calculation (CAFÉ model) has been shown to predict grain structures successfully in various solidification processes [1-3]. Recently, microscopic scale CA models have been developed to simulate microstructures [4-8]. Like phase-field simulations, they can also simulate dendritic morphologies. However, the microscopic scale CA model has lower computational costs. On the other hand, predicting the dendritic grain structures for solidification of samples such as large ingots, billets, and slabs will incur high computational costs. Thus, the macroscopic scale CA model is the most practical and efficient tool to predict solidification grain structures.

According to the conventional model developed by Gandin and Rappaz [1-3], the macroscopic scale CA model coupled with FE heat flow calculations consists of nucleation and growth of the dendritic grain envelope. In this model, the distribution of solute concentration is not considered, thus, the initial solute concentration is given for the whole region. The conditions of nucleation and grain growth are determined by local undercooling obtained from heat flow calculations. To simulate solute segregation,
the distribution of solute concentration must follow the models. Moreover, to simulate and predict macrosegregation, the flow of liquid during solidification needs to be considered [9, 10]. Recently, the CAFÉ model coupled with macrosegregation has been developed and validated by comparing in situ real-time X-ray radiography observations [11-13]. A microscopic scale CA simulation coupled with flow calculation was carried out to have a new mechanism of freckle formation [14]. Natsume and Ohsasa also proposed a macroscopic scale CA model coupled with a finite difference (FD) heat flow calculation (CAFD) model to predict microsegregation in solidification grain structures [15]. In this CAFD model, the effect of fluid flow is considered by an effective partition coefficient. However, this has not been coupled with flow calculation. In the previous work, Natsume developed a numerical model to predict solidification grain structures and macrosegregation based on a macroscopic scale CAFD method coupled with flow calculation of natural convection and shrinkage flow [16, 17]. In the present work, direct simulations of grain growth and macrosegregation using this model were performed to determine the influence of bridging and shrinkage flow on macrosegregation. Also, simulation results of grain structures and macrosegregation under natural convection were demonstrated.

2. Model

In this macroscopic scale CAFD model, nucleation, grain growth, and solute concentration are calculated by the mesoscopic scale model using a cubic CA cell, and heat flow is calculated by the macroscopic scale model using a cubic FD grid. The nucleation adopted the original model proposed by Rappaz and Gandin [18], while a grain growth model based on the octahedral decentered growth algorithm [2, 3] was used. A new method to calculate solute concentration was developed in the previous work [16] while the enthalpy method was used in the heat flow calculation. The flow of liquid is performed in the macroscopic scale model by solving equations (1) and (2).

\[
\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\frac{\mathbf{V} p}{\rho} + \nu \nabla^2 \mathbf{u} - \frac{\nu}{K} \left[ \beta_T (T - T_{ref}) + \beta_c (c - c_{ref}) \right] f_L \mathbf{g} 
\]

\[
\nabla \cdot \mathbf{u} + \beta \frac{\partial f_s}{\partial t} = 0
\]

where \( \mathbf{u} \) is velocity, \( t \) is time, \( p \) is pressure, \( \rho \) is density, \( \nu \) is kinematic viscosity, \( K \) is permeability, \( \beta_T \) and \( \beta_c \) are coefficients of thermal and solutal expansion, respectively, \( T \) is temperature, \( c \) is solute concentration, \( f_L \) and \( f_S \) are fraction of liquid and solid, respectively, \( \mathbf{g} \) is gravity, and \( \beta \) is solidification shrinkage rate. In the permeability in equation (1), an anisotropic permeability model proposed by Natsume et al. [19, 20] was used. In this model, the anisotropic permeability of parallel and normal direction for primary dendrite arms, \( K_P \) and \( K_N \), are given as follows:

\[
K_P = 0.0194 \left( \frac{d_2}{1 + (d_2/d_1)} \right)^2 \frac{f_L^3}{(1 - f_L)^2}
\]

\[
K_N = 0.0097 \left( \frac{d_2}{1 + (d_2/d_1) + (d_2/d_1)^2} \right)^2 \frac{f_L^3}{(1 - f_L)^2}
\]

where \( d_1 \) and \( d_2 \) is the primary and secondary dendrite arm spacing, respectively. In the simulations, the growth of columnar grains was only considered because it was not introduced in the movement of equiaxed grains due to fluid flow. The anisotropic permeability, \( K_P \) and \( K_N \), was set to \( K_x, K_y, \) and \( K_z \) for simplicity.

3. Results and discussion

3.1. Simulations of grain structures and macrosegregation under natural convection

To confirm the effect of natural convection, simulations of unidirectional solidification cooled on the bottom surface of the mold were performed for an Al-10wt% Mg alloy. The size of the calculation domain was 10 x 100 x 80 mm, and the sizes of CA cell and FD grid were 0.25 and 1.25 mm, respectively.
The initial temperature in the entire domain was assumed to be homogeneous at a constant temperature of 640 °C, and the chill was assumed to be set on the bottom of the calculation domain. The temperature and heat transfer coefficient of the chill are always 25 °C and 3000 W/K/m², respectively. Zero-flux boundary conditions were used for temperature and solute concentration on four sides and top surfaces. The anisotropic permeability used was as follows: \( K_x = K_y = K_N \) and \( K_z = K_P \). In this simulation, the solidification shrinkage was not considered.

Figure 1. Grain structure and Mg concentration distribution during solidification.

Figure 1 shows the snapshots of grain structure and distribution of Mg concentration during solidification. Columnar grain structures have formed from bottom to top in alloy melt while Mg-rich plumes rising in the melt were seen due to the subsequent upward flow caused by the thermosolutal buoyant force. Once the plume occurred in the melt above the mushy zone, the morphology of columnar grains varied, and the grains became coarse due to branching of primary and secondary dendrite arms, but it cannot be seen in detail because the grain growth followed the dendritic grain envelope model. Moreover, regions like Mg-rich channels forming in the mushy zone can be observed. Such regions could delay solidification inside the liquid-rich channels and might result in a freckle defect. Similarly, the plume and freckles were observed in the directional solidification of Ga-In alloys under the effect of thermosolutal convection by means of X-ray radioscopy by Shevchenko et al. [21].

Figure 2 shows the average Mg concentration profile of the cross section from bottom to top. The negative segregation occurred in the middle of solidification and the positive segregation occurred at the end of solidification. From these results, the potential of the model to predict macrosegregation and grain structures under natural convection were demonstrated, and it was confirmed that the proposed model was effective in predicting the macrosegregation coupling with the grain structure formation.

Figure 2. The profile of average Mg concentration of cross section at the end of solidification obtained by simulation of unidirectional solidification.

3.2. Simulations of grain structures and macrosegregation with shrinkage flow
To confirm the effect of shrinkage flow, the simulations were performed in the condition wherein macrosegregation formation is possible like the center segregation observed in a continuously cast slab. The original concept of the condition used in the simulation was that of the model experiment of Al-10wt% Cu alloy carried out by Sato et al. [22]. Sato et al. developed a unique mold which can form macrosegregation in the central region of the small ingot, known to be “Sato’s mold” in Japanese research groups. Figure 3 shows the solidification system used in this simulation. The domain size was 50 x 30 x 120 mm, and the sizes of CA cell and FD grid were 0.5 and 2.0 mm, respectively. In a system with the chill shown in Figure 3 (a), the chill was set on the xz surfaces, 70-80 mm from the bottom of the calculation domain, and the xz surfaces other than the chill were set on as the mold surfaces wherein the different heat transfer coefficient were given. The initial temperature in the entire domain was assumed to be homogeneous at a constant temperature of 650 °C. The values of heat transfer coefficient of the chill, mold-1, and mold-2 are 1000 W/K/m², 400 W/K/m², and 200 W/K/m², respectively. Zero-flux boundary conditions were used for solute concentration. The anisotropic permeability used was as follows: \( K_x = K_z = K_N \) and \( K_y = K_P \). For the conditions of simulation, two types of simulation were performed: with and without the chill. In this simulation, natural convection was not considered to determine the influence of shrinkage flow on macrosegregation.

Figure 4 and Figure 5 shows the snapshots of grain structures and distributions of Cu concentration during solidification for a system with the chill, respectively. The bridging of columnar grains formed near the center of the ingot during solidification due to the chill. Molten alloy flowed down through the columnar grains before bridging because of shrinkage flow. In a system without the chill, this phenomenon was not observed. Figure 6 shows the line profile of Cu concentration ratio on centerline at the end of solidification. As shown in Figure 6 (a), the positive segregation was generated in the region below the chill, and the negative segregation was generated in the region above the chill. In a system without the chill, the negative and positive segregations were observed on the upper and lower region, respectively. However, the degree of segregation was weak because the bridging was not formed. Therefore, the primary factor for this macrosegregation was the shrinkage flow with the formation of bridging. From these results, it was confirmed that the shrinkage flow and the bridging of solidification structures played an important role in macrosegregation. Also, in the previous study, it was confirmed that the degree of positive and negative segregation is enlarged by the presence of natural convection. Although the simulated results could not be quantitatively compared with the experimental results by Sato et al. due to the difference in solidification condition, the simulated result could explain the mechanism forming this macrosegregation.
Figure 4. Grain structures during solidification obtained by the simulation with the chill. The elapsed time since solidification start is (a) 15s, (b) 25s, (c) 30s, (d) 35s, and (e) 38s, respectively.

Figure 5. Cu concentration distribution during solidification obtained by the simulation with the chill. The elapsed time since solidification start is 25 sec. (a) shows yz cross section of x = 25 mm, and (b) shows xz cross section of y = 15 mm.

Figure 6. Line profiles of Cu segregation ratio on centerline (x = 25 mm, y = 15 mm) at the end of solidification obtained by the simulations (a) without and (b) with the chill.
4. Conclusions

Two types of direct simulations of grain growth and macrosegregation were performed using a numerical model based on a 3D CAFD method coupled with flow calculation of natural convection and shrinkage flow: (1) the simulation of unidirectional solidification to evaluate the model coupled with natural convection, and (2) simulation for a unique mold proposed by Sato et al.[22] to evaluate the model coupled with shrinkage flow. For the first type, simulations of unidirectional solidification cooled on the bottom surface of the mold were performed for a Al-10wt% Mg alloy. Mg-rich plumes rising in the melt were seen due to the subsequent upward flow caused by the thermosolutal buoyant force, and Mg-rich channels forming in the mushy zone were observed. The columnar grain structures have formed from the bottom to the top in molten alloy, and some grains became coarse after Mg-rich plumes were observed. For the second type, simulations of casting of Al-10wt% Cu alloy for Sato’s mold were performed. The bridging of columnar grains formed during solidification upon cooling, and in the region below it, the positive segregation was generated. Analyzing the simulated results, it was found that the main factor for this macrosegregation is the shrinkage flow with the bridging. In the previous work [16, 17], the influence of the bridging on the macrosegregation was unclear because the simulation without the chill was not performed. In the present work, it has been established that the shrinkage flow and bridging of solidification structures play an important role in the formation of macrosegregation. For future research, the simulation results will be reported in comparing with the experimental data using modified Sato’s mold to evaluate quantitatively the validity of the model in the near future. In addition, a huge amount of computational time will be required to perform 3D flow calculations, especially in large-scale simulations such as continuous casting. A new macroscopic scale CA model coupled with a Lattice Boltzmann method optimized for parallel computing to perform larger-scale simulations will also be reported.

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