A Modified Cellular Automaton Model for the Simulation of Dendritic Growth in Solidification of Alloys

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A modified cellular automaton model (MCA) was developed in order to simulate the evolution of dendritic microstructures in solidification of alloys. Different from the classical cellular automata in which only the temperature field was calculated, this model also included the solute redistribution both in liquid and solid during solidification. The finite volume method, which was coupled with the cellular automaton model, was used to calculate the temperature and solute fields in the domain. The relationship between the growth velocity of a dendrite tip and the local undercooling was calculated according to the KGT (Kurz–Giovanola–Trivedi) model. The effects of constitutional undercooling and curvature undercooling on the equilibrium interface temperature were also considered in the present model. The MCA model was applied to predict the dendritic microstructures, such as the free dendritic growth from an undercooled melt and competitive dendritic growth in practical casting solidification. The simulated results were compared with those obtained experimentally.

KEY WORDS: solidification; microstructural evolution; modified cellular automaton; finite volume method; nucleation; growth kinetics; solute redistribution; diffusion; curvature; columnar dendritic structure; equiaxed dendritic structure; columnar-equiaxed transition.

1. Introduction

Control of solidification microstructures is an important aspect to obtain reproducible and isotropic properties of final casting products in modern casting technology.1) Prediction of microstructure evolution in solidification is a key factor in controlling solidification microstructures. However, it is difficult to predict the structure features because they are determined through the complicated solidification mechanisms, which are controlled by the interplay of thermal, solute and capillary effects, natural or forced convection, and kinetic length or time scales, etc. The recent advances in computer technology and numerical methods have made it possible to analyze transport phenomena (e.g., heat, mass and fluid flow in the mushy zone) to a high level of details.2) The synthesis of that knowledge has, in turn, developed various kinds of deterministic and stochastic models to predict the evolution of microstructures in solidification.

Deterministic models, based on the solution of the continuum equations were first developed for the description of the nucleation, growth and impingement of equiaxed grains in solidification processes.3,4) However, in deterministic approaches, the coupling of the micro model with the conservation equations requires to specify hypothetical grain shapes, such as spheres and cylinders for equiaxed and columnar grains, respectively. Consequently, the deterministic models generally yield an approximation of the position of the solidification front with time, and thus are unable to generate realistic-looking microstructures.

In order to overcome the limitations mentioned above, stochastic models have been developed over the past several years. The first approach initiated by Brown and Spittle5,6) was based on the Monte Carlo (MC) procedure, which were used to predict recrystallization and grain growth in the solid-state phase transformation, as well as to simulate solidification structures in casting.7,8) But there are some drawbacks of the MC methods in simulating solidification structures. For example, they do not explicitly integrate the growth kinetics of the solid–liquid interface. In addition, the MC time step used in the calculations is not correlated with real time. Consequently, they are unable to realistically show the time-evolution of solidification structures.

Rappaz and Gandin9–11) were the first ones applying another stochastic method, cellular automaton (CA) technique, to predict solidification grain structures. This method is based upon the consideration of physical mechanisms on nucleation, growth kinetics of a dendrite tip, and crystallographic orientations. Furthermore, the mechanisms of competitive dendrite growth are directly embedded in the CA algorithm.13) So that the CA model can quantitatively carry out the time-dependent simulation for microstructure evolution, in which the individual grains are identified and their shapes and sizes can be shown graphically. A series of studies using the CA model have been reported by Hong et al. on the simulation of solidification structures in squeeze
casting and planar flow casting,\textsuperscript{12,14} as well as on the prediction of the deflection behavior of columnar grains solidified in a flowing melt.\textsuperscript{15} However, the dendrite growth velocity in the classical CA model mentioned above\textsuperscript{9–15} is referenced only to the local temperature in the solidifying region for a fixed alloy composition. Therefore, the individual grains do not interact directly until they touch each other and it is unable to describe the more detail features such as the side branches and the formation of second phases (eutectic).

On the other hand, phase field models offer the opportunity for better understanding the dynamics of pattern selection and side branch emission in dendritic growth, and they have recently emerged as a viable computational tool for simulating the formation of complex interfacial patterns in solidification.\textsuperscript{16–18} However, phase field models are presently limited to qualitative simulation of a single dendrite or a very small calculation domain due to the large computational capacity needed. In order to study the interactions of dendritic growth in many grains in a solidifying mushy region, a large calculation domain has to be considered, which surely will be a very difficult task in the phase field methods.

Some mesoscopic models for dendritic growth are currently under development.\textsuperscript{19–21} Steinbach and Beckermann et al.\textsuperscript{19,20} used a novel mesoscopic simulation technique to describe the non-steady growth of several equiaxed dendritic grains into a supercooled melt of a pure substance. However, it is clear that their model cannot account for the topology and texture evolution of typical columnar structure. Nastac\textsuperscript{21} has developed a new stochastic model that includes time-dependent calculations for temperature distribution, solute redistribution in the liquid and solid phases, curvature, and growth anisotropy. This model is able to capture the morphological evolution (including the coarsening and growth/branching of the secondary and tertiary dendrite arms) of columnar and equiaxed dendrites, segregation patterns and columnar-to-equiaxed transition (CET) characteristics in casting alloys. Nevertheless, the model cannot identify the different crystallographic orientations since it does not consider the variety of preferential growth direction of nuclei.

The aim of the current study is to develop a new cellular automaton method to model dendritic grain structures in solidification of alloys, covering from microscopic to mesoscopic scales. The present model adopts the same schemes, which have been used in the classical cellular automata, such as the heterogeneous nucleation, the preferential growth orientations of the nuclei and the growth kinetics of a dendrite tip. In addition, it accounts for the curvature, the solute redistribution both in the liquid and solid phases. The present model was applied to simulate the evolution of dendritic structures in solidification of alloys, such as the free dendritic growth, the directional columnar dendritic growth of an organic alloy, and the dendritic growth of Al–Cu alloys solidified directionally and two-dimensionally in a metal mold. The effects of alloy composition, pouring temperature and bulk nucleation parameters on the formation of solidification structures were examined. The computational results were verified by the experimental results.

### 2. Model Description

#### 2.1. Nucleation

Heterogeneous nucleation is assumed to occur on nucleation sites randomly chosen both on the mold surface and in the bulk liquid. Thus, more than one type of nucleation sites was assumed to exist in most microscopic models of solidification, each of these site families becomes active at a critical undercooling.\textsuperscript{9} In the present study, the continuous nucleation model was adopted in which two different Gaussian distributions were considered for describing the heterogeneous nucleation both on the mold wall and in the bulk liquid. The grain density increase \(d_n\) is induced by an increase in the undercooling \(d(\Delta T)\) according to the following Gaussian distribution\textsuperscript{25}:

\[
\frac{d_n}{d(\Delta T)} = \frac{n_{\text{max}}}{\sqrt{2\pi \Delta T_{\sigma}}} \exp \left[ \frac{-1}{2} \left( \frac{\Delta T - \Delta T_{\text{min}}}{\Delta T_{\sigma}} \right)^2 \right] 
\]

where \(\Delta T_{\text{min}}\) is the mean nucleation undercooling, \(\Delta T_{\sigma}\) is the standard deviation, and \(n_{\text{max}}\) is the maximum density of nuclei given by the integral of this distribution from 0 to \(\infty\).

Thus, the density of grains, \(n(\Delta T)\), formed at any undercooling \(\Delta T\), is given by

\[
n(\Delta T) = \int_0^{\Delta T} \frac{d_n}{d(\Delta T')} \, d(\Delta T')
\]

#### 2.2. Growth Kinetics and Orientation

In the present simulation, the growth velocity of a dendrite tip under a certain undercooling was calculated using the KGT (Kurz–Giovanola–Trivedi) model.\textsuperscript{24} The relationships between the growth velocity and the local undercooling for a succinonitrile–acetone alloy and for Al–Cu alloys with various Cu contents are given in Table 1.

The total undercooling at a dendrite tip, \(\Delta T\), is given by the sum of the various contributions to undercooling\textsuperscript{9}:

\[
\Delta T = \Delta T_s + \Delta T_{\sigma} + \Delta T_r + \Delta T_k
\]

where \(\Delta T_s\), \(\Delta T_{\sigma}\), \(\Delta T_r\), and \(\Delta T_k\) are the solutal, thermal, curvature, and kinetic undercooling, respectively. The kinetic undercooling is rather small under normal solidification conditions except for high solidification velocities such as rapid solidification processes.\textsuperscript{9} Therefore, no provision was made for attachment kinetics and only the solutal, thermal and curvature undercoolings were taken into account in the present model.

Nuclei formed on the mold wall or in the bulk liquid were assumed to have random crystallographic orientations within 48 classes. The preferential growth direction corresponds to (10) for cubic metals in the present two-dimen-

### Table 1. Growth kinetics used in the present calculation.

<table>
<thead>
<tr>
<th>Growth velocity (m/sec)</th>
<th>(n(\Delta T) = k_1 \cdot \Delta T_s + k_2 \cdot \Delta T_{\sigma} + k_3 \cdot \Delta T_r + k_4 \cdot \Delta T_k)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al-Cu alloy (mass%Cu=8.0)</td>
<td>(k_1 = 1.1663 \times 10^{-3} \cdot (\text{mass%Cu})^{1.2203}) (k_2 = 5.39996 \times 10^{-6} \cdot (\text{mass%Cu})^{1.3334})</td>
</tr>
<tr>
<td>succinonitrile-1.3 mass% acetone alloy</td>
<td>(k_1 = 8.26 \times 10^{-6}) (k_2 = 8.18 \times 10^{-5})</td>
</tr>
</tbody>
</table>
sional model.

2.3. Solute Redistribution

The assumptions for solute redistribution are made as follows:

1. The local equilibrium at the solid/liquid interface is preserved as follow.

\[ C_i^* = kC_i^* \] ..........................(4)

where \( k \) is the partition coefficient, \( C_i^* \) and \( C_i^* \) are the interface equilibrium concentrations in the solid and liquid phases, respectively.

2. The solute field during solidification is mainly controlled by diffusion in liquid and solid, and no consideration on convective mass transfer is made.

As the solidification proceeds, the solidified cell rejects solute to its neighbor liquid cells. Diffusion within the entire domain is then simulated. The governing equation for the solute redistribution in the liquid region is given by

\[ \frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x} \left[ D_i \frac{\partial C_i}{\partial x} \right] + \frac{\partial}{\partial y} \left[ D_i \frac{\partial C_i}{\partial y} \right] + \frac{C_i(1-k)}{\partial t} \] ..........................(5)

where \( t \) is time, \( D_i \) is the solute diffusion coefficient in the liquid phase, \( f_s \) is the solid fraction, and \( k \) is the partition coefficient. The last term on the right hand side of the Eq. (5) indicates the amount of solute rejected at the solid/liquid interface.

The governing equation for diffusion in solid is given by

\[ \frac{\partial C_i}{\partial t} = \frac{\partial}{\partial x} \left[ D_s \frac{\partial C_i}{\partial x} \right] + \frac{\partial}{\partial y} \left[ D_s \frac{\partial C_i}{\partial y} \right] \] ..........................(6)

where \( D_s \) is the solute diffusion coefficient in the solid phase.

2.4. Macroscopic Thermal Transport

The governing equation for two-dimensional transient heat conduction is given by

\[ \rho C_p \frac{\partial T}{\partial t} = \frac{\partial}{\partial x} \left( \lambda \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( \lambda \frac{\partial T}{\partial y} \right) + \rho \Delta H \frac{\partial f_s}{\partial t} \] ..........................(7)

where \( T \) is the temperature, \( \rho \) is the density, \( C_p \) is the specific heat, \( \lambda \) is the thermal conductivity, and \( \Delta H \) is the latent heat of freezing, respectively.

The boundary conditions at the melt/mold interface and the mold/air interface are given by

Melt/mold:

\[ q = h_{\text{ia}}(T_{\text{mold}} - T_{\text{melt}}) \] ..........................(8)

Mold/air:

\[ q = h_{\text{ia}}(T_{\text{air}} - T_{\text{mold}}) \] ..........................(9)

where \( q \) is the heat flux, \( T_{\text{melt}}, T_{\text{mold}} \) and \( T_{\text{air}} \) are the temperatures of the melt, the mold and the air, and \( h_{\text{ia}} \) and \( h_{\text{ia}} \) are the interfacial heat transfer coefficients at the melt/mold and the mold/air interfaces, respectively.

3. Numerical Method

A numerical model has been developed based on the coupling of the macroscopic heat and mass transport model with a modified cellular automaton model to predict the evolution of dendritic structures in solidification of alloys.

3.1. Nucleation and Growth Algorithm

The cellular automaton model for nucleation and growth simulation consists of (1) the geometry of a cell, (2) the state of a cell, (3) the neighborhood configuration, and (4) several transition rules that determine the state of a given cell during one time step. In this work, the calculation domain is divided into uniform square arrangement of cells. Each cell is characterized by different variables (such as temperature, concentration, crystallographic orientation, solid fraction) and state (e.g. solid or liquid). The cellular automaton evolves in discrete time step, and the state of a cell at a particular time is calculated from the local rule, such as the nucleation and growth kinetics. At the beginning of simulation, the nucleation sites both on the mold surface and in the bulk liquid are randomly set according to the assumptions for solute redistribution.

Once a cell has nucleated, it will grow with a preferential direction corresponding to its crystallographic orientation having a growth velocity determined by the local undercooling and solid concentration. Let us consider a solidified cell labeled “A” which lies at the S/L interface, shown in Fig. 1. There must exist at least one liquid cell within its eight neighbors (including four nearest neighbors and four corner neighbors). Figure 1(a) describes the details of the growth algorithm between the solid cell \( A \) and its liquid neighbor cell. The \( l_j(t_i) \) in Fig. 1(a) is the growth length of the solid cell \( A \) respecting to its liquid neighbor cell \( i \) at time \( t_i \), which can be calculated by

\[ \sum_{j=1}^{N} \left[ \max_{i}(\Delta T(t_i) \times \Delta t_i) \right] = \frac{\sum_{i=1}^{N} \left[ \max_{i}(\Delta T(t_i) \times \Delta t_i) \right]}{\cos \theta} + \sin \theta \] ..........................(10)

where \( \Delta t_i \) is the time step, \( \theta \) is the angle of the cell \( A \)’s preferential growth direction \( \theta_i \) with respect to the linking line between the cell \( A \) and the cell \( i \), and \( N \) indicates the iteration number. \( \nu_i \left( \Delta T(t_i) \right) \) is the growth rate, which can be calculated using the KGT model depending upon the local undercooling, \( \Delta T(t_i) \), taken at the center of the cell \( i \). As mentioned in Sec. 2.2, the present model takes account of the undercooling contributions associated with temperature, composition and curvature. Therefore, the local undercooling \( \Delta T(t_i) \) is given by

\[ \Delta T(t_i) = T_i - C_i(C_i(t_i) - C_i) - m - \frac{T_i(t_i) - \Gamma \theta_i(t_i)}{K_i(t_i)} \] ..........................(11)

where \( T_i \) is the equilibrium liquidus temperature, \( m \) the liquidus slope, \( C_i \) the initial concentration, and \( \Gamma \) the Gibbs–Thomson coefficient. \( K_i(t_i), C_i(t_i) \) and \( T_i(t_i) \) are the mean curvature, the concentration and the temperature of the cell...
on the S/L interface at time $t_n$, respectively. Then, the solid fraction of the cell $i$ at a certain time, $f_s(i)$, can be expressed by

$$f_s(i) = \frac{f_s(t_n)}{L}$$  \hspace{1cm} (12)

where $L$ is the length between the cell A and the cell i, as shown in Fig. 1(a): if i is one of the four nearest east, west, south or north neighbors, $L = dx$; and if i is the corner neighbor, $L = \sqrt{2} dx$. When $f_s(t_n)=1$, which means the growth front of the solid cell A can touch the center of the liquid cell i, the cell i will then transform its state from liquid to solid and get the same orientation index as the cell A.

By means of the algorithm described above, the primary dendrite will grow and coarsen with the preferential $\langle 10 \rangle$ direction, as shown in Fig. 1(b). As the growth and the coarsening of a primary trunk proceed, the solute will be enriched in the liquid near the S/L interface due to the solute redistribution, which will destroy the interface stability and therefore cause the side branching into the secondary arms, as shown in Fig. 1(c).

### 3.2. Calculation of the Interface Curvature

The interface curvature in a cell with the solid fraction $f_i$ is calculated from the following expression,

$$K_i = \frac{1}{dx} \left[ 1 - 2 \frac{f_s(i) + \sum_{j=1}^{n} f_s(j)}{n+1} \right]$$ \hspace{1cm} (13)

where $dx$ is the cell size and $n$ is the number of the neighboring cells. In the present calculation, $n=8$, which includes the surrounding neighbor cells of the first layer. The values of the curvature calculated using Eq. (13) vary from $1/dx$ to 0 for convex surfaces and from 0 to $-1/dx$ for concave surfaces.

### 3.3. Calculation of the Concentration Field

When a cell transforms its state from liquid to solid by nucleation or growth, its concentration will be changed according to Eq. (4). Consequently, this cell will liberate the amount of solute, $dC = C_i^* - kC_i^*$, which is assumed to be distributed to its liquid neighbor cells.
An explicit finite difference scheme was applied for calculating the solute diffusion in both the solid and liquid phases, and the zero-flux boundary conditions were used for the cells located at the surface of the calculation domain. The thermal and physical properties used in the present calculation are listed in Table 2.

### Table 2. Thermal and physical properties used in the present calculation.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Liquidus temperature (K)</td>
<td>928.0 (2 mass% Cu)</td>
</tr>
<tr>
<td>Eutectic temperature (K)</td>
<td>922.0 (4.5 mass% Cu)</td>
</tr>
<tr>
<td>Melting temperature (K)</td>
<td>933.0</td>
</tr>
<tr>
<td>Eutectic concentration (mass%)</td>
<td>33.3</td>
</tr>
<tr>
<td>Partition coefficient</td>
<td>0.17 (Al-Cu alloy)</td>
</tr>
<tr>
<td>m</td>
<td>0.1 (succinonitrile-acetone alloy)</td>
</tr>
<tr>
<td>Slope of the liquidus line (K/mass%)</td>
<td>-3.36 (Al-Cu alloy)</td>
</tr>
<tr>
<td>Specific heat (J/kg·K)</td>
<td>1086 (Al-Cu alloy)</td>
</tr>
<tr>
<td>Density (kg/m³)</td>
<td>2780 (Al-Cu alloy)</td>
</tr>
<tr>
<td>Thermal conductivity (W/m·K)</td>
<td>192.5 (Al-Cu alloy)</td>
</tr>
<tr>
<td>Volumetric latent heat (J/m³)</td>
<td>1.107 × 10⁶</td>
</tr>
<tr>
<td>Mold/casting interfacial heat transfer coefficient (W/m²K)</td>
<td>1000</td>
</tr>
<tr>
<td>Mold/air interfacial heat transfer coefficient (W/m²K)</td>
<td>50</td>
</tr>
<tr>
<td>Solute diffusion coefficient in liquid (m²/sec)</td>
<td>10⁻⁸ (Al-Cu alloy)</td>
</tr>
<tr>
<td></td>
<td>1.3 × 10⁻⁶ (succinonitrile-acetone alloy)</td>
</tr>
<tr>
<td>Solute diffusion coefficient in solid (m²/sec)</td>
<td>10⁻¹² (Al-Cu alloy)</td>
</tr>
<tr>
<td></td>
<td>1.3 × 10⁻¹⁰ (succinonitrile-acetone alloy)</td>
</tr>
</tbody>
</table>

An explicit finite difference scheme was applied for calculating the solute diffusion in both the solid and liquid phases, and the zero-flux boundary conditions were used for the cells located at the surface of the calculation domain. The thermal and physical properties used in the present calculation are listed in Table 2.

#### 3.4. Solution Scheme for Macroscopic Thermal Transport

In order to analyze macroscopic transient heat transfer problems, the calculation domain was divided into control volumes. The governing equations and boundary conditions shown in Eqs. (7) to (9) were solved by the explicit finite difference algorithm.

#### 3.5. Coupling of the Finite Volume Method and the Modified Cellular Automaton Model

The present model consists of two schemes: the modified cellular automaton model (MCA) for simulating the evolution of dendritic structures and the finite volume method for evaluating the macroscopic heat and solute transport in solidification. Figure 2 indicates the schematic drawing of the calculation domain with control volumes (CV) which are used for the calculation of macroscopic heat transfer. Each CV consists of 30 × 30 square cellular automaton (CA) cells for the MCA calculation. As the thermal diffusivity of metallic alloys is 3–4 orders of magnitudes greater than the solute diffusivity, the kinetics for microstructure evolution can be assumed to be solute transport-controlled, and therefore the thermal diffusion can be considered to be complete in the microscopic scale.

Thus, for the sake of simplicity and the reduction of calculation time, the temperatures of all cells within a CV are assumed to be uniform. Based on the calculated temperature profile in the cells, the calculation of nucleation and growth as well as the solute redistribution are carried out by the MCA model as described above.

In order to reduce the computational time, two different time steps were used; one for macroscopic heat transfer cal-
calculation based on CV, and the other for microscopic MCA calculation based on CA cells, as follows:

**CV time step:**

\[
\delta t_{CV} = \frac{\Delta X_{CV} \times \rho C_p}{4.5 \times \lambda} \quad \text{(14)}
\]

where \(\Delta X_{CV}\) is the CV size, \(\rho C_p\) is the volumetric specific heat, and \(\lambda\) is the thermal conductivity.

**MCA time step:**

\[
\delta t_{MCA} = \frac{1}{4.5} \min \left[ \frac{dx}{V_{\max}}, \frac{dx^2}{D_l} \right] \quad \text{(15)}
\]

where \(V_{\max}\) is the maximum growth velocity obtained by scanning the growth velocities of all interface liquid cells during each time step and \(D_l\) is the solute diffusion coefficient in liquid.

When a liquid cell solidifies by nucleation or growth, it will release the latent heat of freezing, which is evaluated by

\[
\Delta T_i = \frac{\Delta H_V \times dx^2}{\rho C_p \times \Delta X_{CV}^2} \quad \text{(16)}
\]

where \(\Delta T_i\) is the equivalent temperature recovered due to the latent heat released by one solidified cell and \(\Delta H_V\) is the volumetric latent heat of freezing. The temperature of the CV node is then recovered based on Eq. (16) by the newly solidified cells included in this CV. Using these updated temperatures, macroscopic heat transfer calculation can be continued. This series of calculation will be repeated until the end of solidification. The flowchart of the present calculation is shown in Fig. 3.

**4. Results and Discussion**

**4.1. Free Dendritic Growth in an Undercooled Melt**

In order to simulate free dendritic growth into an undercooled melt, the calculating domain is divided into 320 \times 320 cells with a cell size of 0.2 \(\mu\)m, which is fine enough to resolve a dendrite tip radius. In the beginning of simulation, one nucleus with the preferential growth orientation of 0° or 45° with respect to the horizontal direction was assigned in the center of the area. The initial concentration of the calculation domain was assumed to be \(C_0\).

The simulated dendrite morphology of an Al–2.0mass% Cu alloy solidified into an undercooled melt (\(\Delta T=40\) K) is shown in Fig. 4 for three stages: (a) the initial growth stage; (b) the initiation of the side branch emission; and (c) the dendrite with well-developed side branches (calculation domain: 320 \times 320; cell size: 0.2 \(\mu\)m).

**Fig. 3.** Flowchart of the MCA-FVM coupling and calculation procedure.

**Fig. 4.** The simulated dendritic shapes during the isothermal growth of an Al–2.0mass%Cu alloy at 888 K (\(\Delta T=40\) K): (a) the initial growth stage; (b) the initiation of the side branch emission; and (c) the dendrite with well-developed side branches (calculation domain: 320 \times 320; cell size: 0.2 \(\mu\)m).
tration profiles in both the solid and liquid phases. Within the solid region, along the centerline of primary trunks or side arms, there exists a spine with lower concentration, which is considered as the result of the combined effect of curvature and interface kinetics. The concentration in solid near the solid/liquid interface shows the higher concentration where the final solidification occurs. It could be seen from Fig. 4(c) that the tertiary arm branching occurs only at one side of the secondary arms. These interesting phenomena have been observed in the previously reported experiments and are also consistent with simulation results by the phase field models. Figure 4 illustrates the capability of the present model to nicely depict the dendrite evolution features, including the growing and coarsening of the primary trunk, the branching of the secondary and tertiary dendrite arms, as well as the solute segregation patterns.

It can be noted that the present MCA model has some significant advantages compared with phase field models. Firstly, the MCA model has the excellent computational efficiency and only needs a small memory size. For example, the computational time for Fig. 4 was only about 4 min on a Pentium PC-450 MHz and the memory size needed was only about 10MB. Whereas it is well known that phase field models suffer from a low computational efficiency and a very large memory resource. In case of the phase field models, it usually takes about a couple of days to calculate a single dendrite formation using a Pentium-level personal computer. Secondly, phase field models have a serious cell size limitation such as in the order of $10^{-8}$ m due to the limiting condition on the interface thickness. On the other hand, a relatively wide range of cell size such as $10^{-8}$–$10^{-5}$ m is available in the MCA model, which enables it to simulate the evolution of solidification structures in practical casting solidification. And thirdly, in case of phase field models, the side branching into the secondary arms around the dendrite tip is known to be possible only under the existence of artificial noise source. However, the MCA model can carry out the simulation of dendrite growth with the well-developed secondary and tertiary arms in the very natural way without imposing any artificial noises.

4.2. Prediction of the Structures Formed in the Directional Solidification

4.2.1. Grain Selection in the Columnar Dendritic Growth

As reported by Esaka, in situ observation of the dendritic solidification of transparent organic materials is a very powerful tool for the study of the grain selection or competitive growth in the columnar zone. Figure 5 indicates the competition between three columnar grains of a succinonitrile–1.3mass% acetone alloy solidified directionally with a constant thermal gradient, $G=1.900$ K/m, and a velocity of the liquidus isotherm, $8.6\times10^{-5}$ m/s: (a) the experimental result and (b) the simulation result by the present MCA model. The calculation domain consists of 760 $\times$ 1 500 CA cells and the cell size was 5 $\mu$m. In the beginning of the simulation, three kinds of nuclei were assigned at the bottom of the calculation domain, having their (10) crystallographic orientation at about 6° (left), 40° (center), and 5° (right), respectively. The initial undercooling was chosen to be 2 K to control the growth velocity of a dendrite tip. The grain selection or competitive growth among columnar grains can be easily seen from the figure. It is obvious from Fig. 5 that the predicted dendrite pattern is in good agreement with that obtained experimentally.

4.2.2. Prediction of Dendritic Structures in Directional Solidification of Al–Cu Alloys

The present model was applied to predict the evolution of dendritic grain structures of Al–2.5mass%Cu and Al–4.5mass%Cu alloys unidirectionally solidified over a copper chill plate with a constant temperature of 298 K. The nucleation parameters in Eq. (1), $\eta_{\text{max}}, \Delta T_{\text{sat}}, \text{ and } \Delta T_{\text{cur}}$, are listed in Table 3. The symbols indexed “s” and “b” are corresponding to nucleation parameters on the mold surface and in the bulk liquid, respectively.

Figure 6 indicates the simulated and experimental macro- and micro-structures of Al–Cu alloys unidirectionally solidified with a pouring temperature of 1 013 K. The figures on the upper row indicate the simulated structures and on the bottom row the experimental ones. The figures on the left column indicate the case of the Al–2.5mass%Cu and the right for the case of the Al–4.5mass%Cu. Figures 6(a) and 6(c), indicating the grain structures simulated by a classical cellular automaton model, are in good agreement with (e) and (g) obtained experimentally. For this simulation, the casting domain was divided into 1 000 $\times$ 2 000 cells with a cell size of 30 $\mu$m. Figures 6(b) and 6(d), indicating the dendritic structures simulated by the present MCA model, are also in good agreement with (f) and (h) obtained experimentally. For this simulation, a calculation domain was divided into 400 $\times$ 400 cells with a cell size of 3 $\mu$m. It is seen from the figures that the present MCA model can be applied to predict both the columnar and equiaxed dendritic growth morphology in casting solidification.

4.3. Prediction of Grain Structures Solidified in a Rectangular Steel Mold

The present model was also applied to predict the den-
The dendritic structure evolution of an Al–2.5 mass% Cu alloy solidified in a rectangular steel mold under the time-depended temperature boundary condition. The size of the mold cavity was 2 cm x 1 cm x 0.1 mm, and 1/4 area of the mold cavity was chosen for the simulation because of its geometrical symmetry. The solidification was considered to be two-dimensional problem since the bottom and top surfaces were insulated. The mold was 4 cm in thickness and its initial temperature was 298 K. As shown in Fig. 2, the whole domain including the mold and the casting was divided into control volumes for macroscopic heat transfer calculation, and the control volumes within the casting domain were further divided into CA cells for simulating the microstructural evolution. In this case, each control volume consists of 30 x 30 CA cells and the cell size was taken to be 10 μm, which is considered small enough to simulate the side branching in dendritic growth.

In practical castings, there usually are three kinds of grain structures formed: chill zone, columnar zone, and equiaxed zone. The different structures are determined by various process variables. Generally, the pouring temperature of melt has a significant effect on the structure of casting. The length of the columnar zone increases with an increase of pouring temperature. The mechanism proposed is that for a low pouring superheat, new crystals may rapidly form in the thermally undercooled region near the mold.

### Table 3. The nucleation parameters used in the present calculation.

<table>
<thead>
<tr>
<th>Figure</th>
<th>Material</th>
<th>$n_{\text{ave}}$ (m$^{-3}$)</th>
<th>$\Delta T_{\text{ave}}$ (K)</th>
<th>$\Delta T_{\text{p}}$ (K)</th>
<th>$n_{\text{ave,p}}$ (m$^{-3}$)</th>
<th>$\Delta T_{\text{p}}$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fig. 6</td>
<td>Al-2.5 mass% Cu</td>
<td>1.8 x 10$^9$</td>
<td>0.1</td>
<td>8.0 x 10$^4$</td>
<td>5.0</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>Al-4.5 mass% Cu</td>
<td>1.8 x 10$^9$</td>
<td>0.1</td>
<td>1.6 x 10$^4$</td>
<td>2.0</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>(a)</td>
<td>7.0 x 10$^8$</td>
<td>0.1</td>
<td>8.0 x 10$^4$</td>
<td>7.0</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>(b)</td>
<td>1.8 x 10$^3$</td>
<td>0.1</td>
<td>1.6 x 10$^4$</td>
<td>4.0 x 10$^3$</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>(c)</td>
<td>2.2 x 10$^3$</td>
<td>0.1</td>
<td>4.0 x 10$^3$</td>
<td>3.5</td>
<td>0.1</td>
</tr>
<tr>
<td>Fig. 7</td>
<td>(a)</td>
<td>7.0 x 10$^8$</td>
<td>0.1</td>
<td>8.0 x 10$^4$</td>
<td>7.0</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>(b)</td>
<td>1.8 x 10$^3$</td>
<td>0.1</td>
<td>1.6 x 10$^4$</td>
<td>3.5 x 10$^3$</td>
<td>0.1</td>
</tr>
<tr>
<td></td>
<td>(c)</td>
<td>2.2 x 10$^3$</td>
<td>0.1</td>
<td>4.0 x 10$^3$</td>
<td>3.0</td>
<td>0.1</td>
</tr>
</tbody>
</table>

Note: the relationships between the nucleus densities for 3D, $n_{\text{ave}}$ (m$^{-3}$) and $n_{\text{ave,p}}$ (m$^{-3}$), and the ones for 2D, $n_{\text{ave,3}}$ (m$^{-3}$) and $n_{\text{ave,2}}$ (m$^{-3}$) are given by:

$$n_{\text{ave,p}} = \frac{\pi}{4} (n_{\text{ave,3}})^2$$

$$n_{\text{ave,2}} = \left(\frac{\pi}{6}\right) (n_{\text{ave,3}})^{3/2}$$

Fig. 6. Comparison between predicted and experimental results in directional casting of Al-Cu alloys with a pouring temperature of 1 013 K. (a), (b), (c) and (d) indicate the simulated results; and (e), (f), (g) and (h) indicate the experimental results. (a) and (c) indicate the macrostructures obtained by a classical CA model, and (b) and (d) indicate the microstructures obtained by the MCA model.
wall and move into the bulk liquid by convection and turbulence. However, this so-called the big-bang nucleation could be eliminated by increasing the pouring superheat. Thus, the different conditions for bulk nuclei generation were implemented. The number of nucleation density decreases and the amount of undercooling increases with an increase of the pouring temperature. The nucleation parameters used for the simulations are listed in Table 3.

Figure 7 shows the evolution of solidification microstructures: (a), (b) and (c) indicate the simulation results; and (d), (e), and (f) indicate the experiment results. (a) and (d): fully columnar structures with $T_{\text{pour}}=1093$ K; (b) and (e): mixed structures (CET) with $T_{\text{pour}}=983$ K; and (c) and (f): fully equiaxed structures with $T_{\text{pour}}=933$ K.

The simulation results by the present MCA model were compared with those by the classical CA model in which no consideration on the solute redistribution and transport during solidification was made. Figure 8 indicates the simulation results by the classical CA model. Comparing Fig. 8 with Fig. 7, it is readily apparent that the present MCA model is able to nicely depict the dendrite features in de-
tails, whereas the classical CA model can only display the rough morphology of grain structures. In addition, it is also noted that the bulk nucleation undercooling for obtaining equiaxed grain structures should be a little smaller for the classical CA model than the present MCA model. This is because in the classical CA model no consideration on solute redistribution was made, and the concentration in the remained liquid is kept constant. On the other hand in the present MCA model, the liquid concentration in the S/L interface will increase as the dendrites grow, leading to the decrease of the local undercooling according to Eq. (11). Therefore, the growth velocity of a dendrite tip in the classical CA model is faster than that in the present MCA model.

5. Conclusion

A new comprehensive two-dimensional cellular automaton model, which is based upon the coupling of a modified cellular automaton model with the finite volume method, has been developed in order to simulate the evolution of dendritic structure in solidification of alloys. The present MCA model includes the nucleation and growth kinetics, the preferred growth orientation of a dendrite tip, as well as the constitutional undercooling and curvature undercooling. The heat transfer and solute redistribution including diffusion in solid and liquid are solved by the finite volume method. It was found that the present model can quantitatively describe the evolution of dendritic growth features, including the growing and coarsening of the primary trunks, the branching of the secondary and tertiary dendrite arms, as well as the solute segregation patterns. The free dendritic growth from an undercooled melt can be modeled by the present model, which was very similar to those by the phase field models. Moreover, the present MCA model has significant advantages compared to the phase field models, such as its excellent computational efficiency and no cell size limitation. The present MCA model can predict not only the evolution of solidification grain structures, but also dendritic growth morphology inside the grains, whilst the classical cellular automata can only predict the rough grain structures. The three kinds of typical structures: the fully columnar dendritic structure, the fully equiaxed dendritic structure and the columnar to equiaxed transition, can be satisfactorily predicted by the present model, and the predicted microstructures are in good agreement with those obtained experimentally. Accordingly, it can be concluded that the present MCA model is able to act as an applicable bridge over the gap between the microscopic dendritic growth simulation and the mesoscopic multi-dendritic growth simulation in practical casting.

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REFERENCES