Prediction of Solidification Microstructure and Columnar-to-equiaxed Transition of Al–Si Alloy by Two-dimensional Cellular Automaton with “Decentred Square” Growth Algorithm

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A Cellular Automaton (CA) -Finite Difference (FD) coupling model was developed to analyze the evolution of solidification microstructure and the columnar-to-equiaxed transition (CET) in Al–Si alloy. Kobayashi's microsegregation equation was adopted to describe the solute diffusion in solid phase, and a “decentred square” growth algorithm with coordinate transformation was performed to describe the grain growth and the entrapment of neighbor cells. Through the examination on the effects of operation parameters and nucleation parameters on solidification morphologies, it was found that the length of columnar grains is controlled by the dendrite tip growth kinetics, and that the width of columnar grains is controlled by the implicit relationship between nucleation density and cooling rate at ingot surface. It was also found that the size of equiaxed grains is controlled by the competition of the nucleation and the grain growth. With the controllability of nucleation density in the bulk of liquid for equiaxed grain size, the nucleant and the nucleation density in actual Al–Si alloy were estimated. Both of the CET criteria based on the solidification path by CA-FD coupling model and the one based on the curves of critical temperature gradient conditions by Hunt's model were strongly dependent on nucleation undercooling and Si concentration. A good agreement was obtained between these two.

KEY WORDS: Al–Si alloy; solidification microstructure; grain size; nucleant; CET; Cellular Automaton.

1. Introduction

Cellular Automaton (CA) models have been developed for the prediction of dendritic grain structures formed in solidification processes. Rappaz and Gandin¹⁾ have firstly proposed the CA model for the prediction of solidification microstructure in uniform temperature field. Its main idea was to track the positions of growth fronts of the dendritic grains, however, the internal structure of the dendritic grains was not modeled. In other words, only the development of grain envelopes was taken into account by using the dendrite tip growth kinetics related to the local undercooling. Since then, the model has been developed to predict the solidification microstructure not only in uniform temperature field but also in gradient temperature field,^{2,3)} from two dimensional configuration to three dimensional one. A fully coupling between finite element (FE) method and cellular automaton model (CA)⁴⁾ was realized at last. Recent CA models also considered the effects of fluid flow and macrosegregation.⁵⁾

To meet the demands of industrial applications, CA model is usually coupled with finite difference (FD) or finite element (FE) method. At the macroscopic scale, the temperature field is resolved by using FD or FE method. Thus the local undercooling obtained in FD mesh nodes is

related to the nucleation and the dendrite tip growth kinetics in CA cells at microscopic scale. Cho *et al.*⁶⁾ have coupled FD with CA model in order to predict the solidification microstructure of Al–Si alloy and examined the effects of nucleation parameters. Scheil's equation for microsegregation was combined with heat transfer procedure in their model. Nakajima and Mizoguchi,⁷⁾ adopting Kobayashi's equation for microsegregation⁸⁾ which considers the solute diffusion in solid phase, predicted the temperature field and the solidification process for multi-component alloy.

In the present work, a CA-FD coupling model is developed to analyze the effects of operation parameters and nucleation parameters on the development of solidification microstructure in Al–Si alloy. The microsegregation combined with heat transfer procedure in FD method is described by Kobayashi's equation, and the grain growth procedure in CA model is described by two dimensional (2D) “decentred square” growth algorithm.³⁾ The effects of operation parameters and nucleation parameters on the solidification microstructure, especially on the size of columnar and equiaxed grains, are discussed by comparing predicted results with previous experimental ones. Identification of nucleant and estimation of its nucleation density in actual Al–Si alloy is done using the CA-FD coupling model. In addition, the critical conditions for columnar-to-equiaxed

transition (CET) are predicted from the development of solidification microstructure using the CA-FD coupling model, and are compared with those from the models of Hunt and Stefanescu.

2. CA-FD Coupling Model

An unsteady two dimensional heat transfer procedure combined with solute redistribution at solid/liquid (S/L) interface during solidification is considered in Al-Si alloy ingot. The accurate temperature field is obtained following the energy balance. The temperature is then interpolated into CA cells and related to the nucleation procedure and the grain growth procedure.

2.1. FD Method for Temperature Field

The heat transfer equation combined with microsegregation model^{7,8)} is adopted to obtain the temperature field. The unsteady heat transfer equation is expressed as

$$\frac{\partial H}{\partial t} = \nabla \cdot (\kappa \nabla T) \dots\dots\dots(1)$$

It is rewritten by the explicit difference scheme as

$$H_{n+1} = H_n + \nabla \cdot (\kappa \nabla T_n) \Delta t \dots\dots\dots(2)$$

where H_{n+1} is the enthalpy per unit volume at $n+1$ -th time step, κ is the thermal conductivity, T_n is the temperature at n -th time step, Δt is the time step in FD method which is determined by stability analysis of explicit difference scheme. The boundary condition adopted at ingot surface is

$$-\kappa \frac{\partial T}{\partial n} = h(T - T_\infty) \dots\dots\dots(3)$$

where n is the normal to ingot surface, h is the heat transfer coefficient, T_∞ is the surrounding temperature.

The energy balance among enthalpy, H , solid fraction, f_s , and temperature, T , is expressed in Eq. (4). Assuming the molten alloy being a dilute solution and the local equilibrium at solid/liquid (S/L) interface, temperature, T_{n+1} , is obtained through Eq. (5). The microsegregation model adopted is Kobayashi's solution of Brody-Fleming's solidification model, which considers solute diffusion in solid phase.⁸⁾ Here solute concentration in liquid, C_{n+1} , is expressed as a function of solid fraction, $f_{S,n+1}$, as shown in Eq. (6).

$$H_{n+1} = \rho \Delta H_{LS} (1 - f_{S,n+1}) + \rho C_p T_{n+1} \dots\dots\dots(4)$$

$$\frac{1}{T_{n+1}} = \frac{1}{T_L^0} + \frac{R}{\Delta H_{LS}} (1 - k^{S/L}) \frac{C_{n+1}}{W} \dots\dots\dots(5)$$

$$C_{n+1} = C_n \left(\frac{P_{n+1}}{P_n} \right)^\zeta \left[1 + \frac{k^{S/L} (1 - k^{S/L}) \beta^3}{2\gamma (1 - \beta k^{S/L})^3} (Q_{n+1} - Q_n) \right] \dots\dots\dots(6)$$

where ρ is the density, C_p is the specific heat. T_L^0 is the melting point of pure metal, ΔH_{LS} is the latent heat of solidification as a function of solute concentration. C^0 and W are

the atomic weight of solute element. $k^{S/L}$ is the equilibrium partition coefficient. The terms of P_n , Q_n , β , γ and ζ are expressed as functions of solid fraction, $f_{S,n}$, secondary arm spacing, λ_2 , and diffusion coefficient in solid phase, D_S .⁸⁾

Given the initial conditions of solute concentration, C^0 , superheat, $\Delta T_{\text{superheat}}$, and external cooling condition (heat transfer coefficient, h), temperature, T_{n+1} , solid fraction, $f_{S,n+1}$, and solute concentration, C_{n+1} , can be determined simultaneously by iterative solution of Eqs. (2), (4), (5) and (6).

Once the concentration reaches the eutectic point, the temperature and the concentration are maintained until the solid fraction reaches unity. Afterwards, they change again with solidification progress.

2.2. CA Model for Solidification Microstructure in Gradient Temperature Field

The final solidification microstructure observed in cross section of an ingot can be characterized by three main phenomena: local undercooling, nucleation of grains and its preferential growth orientation. The CA model including the nucleation procedure and the grain growth procedure is adopted to predict the development of solidification microstructure in a gradient temperature field.

2.2.1. Local Undercooling

The temperatures in FD mesh nodes are interpolated into CA cells linearly firstly in time and then in space, because both time step and cell size in CA procedure are smaller than those in FD procedure.

The micro time step, δt , is determined to limit the interface moving within one cell in one time step,

$$\delta t = \min \left[\frac{l_{CA}}{v_{\text{tip}}(\Delta T_{\text{max}})}, \Delta t \right] \dots\dots\dots(7)$$

where l_{CA} is CA cell size, $v_{\text{tip}}(\Delta T_{\text{max}})$ and ΔT_{max} are the maximum growth rate of dendrite tip and the maximum local undercooling among all growing cells found in the calculation region, respectively.

The CA cell size is set as 1/4 or 1/5 length of primary arm spacing, λ_1 , which is determined from the cooling rate at ingot surface, $\dot{T}_{\text{surface}}$ (in Table 1).

Firstly the temperature of a FD mesh node at CA time, $t + \delta t$, is obtained by linear interpolation of the corresponding values at time, t and $t + \Delta t$. Then the temperature of a CA cell is obtained by double linear interpolation of the corresponding values at four nearest FD mesh nodes.

Once the temperature at CA cell is lower than liquidus temperature, it is undercooled and becomes the potential cell for the nucleation and the grain growth.

2.2.2. Nucleation

Both locations and crystallographic orientations for new nucleus are chosen randomly. The continuous nucleation distribution, $dn_V/d(\Delta T_V')$, is described by Gaussian distribution (Eq. (8)). When the nucleation probability of a CA cell, $P_V = \delta n_V \cdot V_{CA}$, is greater than a random number, r ($0 \leq r \leq 1$), during a micro time step, δt , the cell is nucleated. Its state is changed from liquid to solid, and it is given a random crystallographic orientation, θ , which is the angle

Table 1. Thermophysical properties of Al-Si alloys used in the CA-FD coupling model.

Parameter	Symbol	Unit	Al-3mass%Si	Al-5mass%Si	Al-7mass%Si
Initial solute concentration in liquid	C^0	-	0.03	0.05	0.07
Melting point of pure Al	T_L^0	$^{\circ}\text{C}$		660.35 ⁹⁾	
Liquidus temperature	T_L	$^{\circ}\text{C}$	642.816	632 ¹¹⁾	618 ⁹⁾
Initial temperature	T_{ini}	$^{\circ}\text{C}$		800	
Primary arm spacing	λ_1	m		$220 \times 10^{-6} \times (\dot{T}_{S, surface})^{-0.55}$ ¹¹⁾	
Secondary arm spacing	λ_2	m		$5 \times (M \cdot t_{SL})^{1/3}$ ¹¹⁾	
Coefficient in the expression of λ_2	M	m^3/s	7.73×10^{-18} ¹¹⁾	6.30×10^{-18} ¹¹⁾	5.43×10^{-18} ¹¹⁾
Solidification time	t_{SL}	s	(local time – solidification beginning time)		
Diffusion coefficient of Si in solid phase	D_S	m^2/s	$D_S = D_0 \cdot \exp(-Q/RT)$, $T, [K]$ ¹²⁾		
Diffusion constant in the expression of D_S	D_0	m^2/s		2.0×10^{-4} ¹²⁾	
Activity energy	Q	J/mol		1.335×10^5 ¹²⁾	
Specific heat	C_p	J/m ³ K		2.6×10^6 ¹³⁾	
Thermal conductivity	κ	W/m K	$\kappa = \kappa_S \cdot f_S + \kappa_L \cdot f_L$		
Thermal conductivity in liquid	κ_L	W/m K	$\kappa_L = 41.5 + 0.0312T$ ¹⁴⁾ $T, [K]$	90 ¹¹⁾	$\kappa_L = 365 + 0.028T$ ¹⁴⁾ $T, [K]$
Thermal conductivity in solid	κ_S	W/m K	$\kappa_S = 253 - 0.110T$ ¹⁴⁾ $T, [K]$	104 ¹¹⁾	$\kappa_S = 233 - 0.110T$ ¹⁴⁾ $T, [K]$
Density	ρ	kg/m ³		2452.5 ⁹⁾	
Partition coefficient	$k^{S/L}$	-	$k^{S/L} = 0.1125 + 0.25 \times C_{Si}$		
Latent heat of solidification	ΔH_{LS}	kJ/kg	$397.0 - 473.68C_{Si} - 2004.5C_{Si}^2$		
Gibbs-Thomson coefficient	Γ	K m	1.96×10^{-7} ¹⁴⁾	1.96×10^{-7}	1.96×10^{-7} ¹⁴⁾
Slope of liquidus	m	K/-	-600. ¹⁴⁾	-625.	-650. ¹⁴⁾
Diffusion coefficient of Si in liquid phase	D_L	m^2/s	7.0×10^{-9} ¹⁴⁾	6.725×10^{-9}	6.45×10^{-9} ¹⁴⁾

Fig. 1. Schematic diagram of 2D “decentred square” cellular automaton growth algorithm for “decentred square” growth of cell v and the entrapment of its neighbor cell μ .³⁾ The symbols denoted by $\circ$ (hollow circles) correspond to the centers of cells in the CA network, and the symbols denoted by $\bullet$ (solid circles) are their corresponding virtual growth centers. At a certain time, the growth envelope of cell v (the larger hollow square envelope centered at C_v) has entrapped the neighbor cell μ . The sequent initial growth length of cell μ , L_{μ}^t , is a half side length of the truncated gray square. And the virtual growth center of cell μ is at site C_{μ} which biases the center of cell μ .

of one dendrite arm (*i.e.* [10] direction) with respect to the CA x -axis. The angle is among $(-\pi/4, \pi/4)$ from the view of the four-fold symmetry of dendritic grain. Here, V_{CA} is the volume of a CA cell, δn_v is the increase of grain density corresponding to undercooling increase, $d(\Delta T_v)$,

$$\frac{dn_v}{d(\Delta T_v')} = \frac{n_v}{\sqrt{2\pi} \cdot \Delta T_{v,\sigma}} \exp \left[-\frac{1}{2} \left(\frac{\Delta T_v' - \Delta T_v}{\Delta T_{v,\sigma}} \right)^2 \right] \dots (8)$$

$$\begin{aligned} \delta n_v &= n_v [\Delta T_v + \delta(\Delta T_v)] - n_v (\Delta T_v) \\ &= \int_{\Delta T_v}^{\Delta T_v + \delta(\Delta T_v)} \frac{dn_v}{d(\Delta T_v')} d(\Delta T_v') \dots (9) \end{aligned}$$

where n_v is the maximum nucleation density formed in the bulk of liquid. ΔT_v and $\Delta T_{v,\sigma}$ are the maximum undercooling and the standard deviation of the nucleation distribution, respectively. A similar procedure is applied to the heterogeneous nucleation of grains at ingot surface. Thus, two distributions are used for the heterogeneous nucleation in the bulk of liquid ($\Delta T_v, \Delta T_{v,\sigma}, n_v$) and at ingot surface ($\Delta T_s, \Delta T_{s,\sigma}, n_s$). The nucleation densities n_v [m^{-3}] and n_s [m^{-2}] are applied to three-dimensional geometries. The following stereological relationships were used to deduce the corresponding values n_v^* [m^{-2}] and n_s^* [m^{-1}] for the two-dimensional (2D) cellular automaton calculations: $n_v = \sqrt{\pi/6} \cdot (n_v^*)^{3/2}$ and $n_s = \pi/4 \cdot (n_s^*)^2$.¹⁾

2.2.3. Grain Growth

Gandin and Rappaz¹⁻³⁾ has developed three kinds of grain growth algorithms. One is “square” growth al-

gorithm¹⁾ in which both nucleation grains and entrapped grains grow in a square envelope in two-dimensional (2D) case. It can only be applied to uniform temperature field. Since the grain orientation is strongly biased by the artificial anisotropy of CA mesh system, “dendrite tip correction” should be done to keep the original orientation of grains. The subsequent one is “rectangle” growth algorithm²⁾ in which the nucleation grains grow in a square envelope while the entrapped grains grow in a rectangular envelope in 2D case. It can maintain the original grain orientation without “correction” and can be applied to non-uniform temperature field, but it is difficult to extend to three-dimensional (3D) case. Thus comes out “decentred square” growth algorithm³⁾ in which both the nucleation grains and the entrapped grains grow in a square envelope in 2D case. In the algorithm, the square envelope is always truncated to a proper size to avoid overgrowth and thus the virtual growth center for the cell usually biases its center in CA network. The “decentred square” growth algorithm can maintain the original grain orientation, describe more exactly the development of solidification microstructure in non-uniform temperature field and is easy to develop in 3D case. So it was chosen in the present work.

In 2D CA network, separated by a distance, l_{CA} , as shown in **Fig. 1**, a growing cell v is denoted by several variables such as state (which is set as solid), crystallographic orientation, θ , virtual growth center, C_v (*i.e.* a center of square growth envelope), and growth length, L_v^t (*i.e.* half side length of square growth envelope). Assuming a local uniform temperature in the scale of a cell, growth length, L_v^t , is given by

$$L_v^t = L_v^{t-\delta t} + \frac{v_{\text{tip}}(\Delta T_v^{t-\delta t}) \cdot \delta t}{\sqrt{2}} \dots\dots\dots(10)$$

where v_{tip} is the growth rate of dendrite tip. For Al–Si alloy, the dendrite tip growth kinetics can be calculated by the following approximate expression which fits well with the predictions of KGT model over a large range of undercoolings,

$$v_{\text{tip}}(\Delta T) = A \cdot \Delta T^n \dots\dots\dots(11)$$

where n is 2.7, A is 1.7×10^{-5} m/s/K^{*n*}, 5.75×10^{-6} m/s/K^{*n*} and 2.9×10^{-6} m/s/K^{*n*} for Al–3mass%Si, Al–5mass%Si, and Al–7mass%Si alloy, respectively.^{1,9)} Here, the values of coefficients n and A for Al–3mass%Si and Al–7mass%Si alloys were proposed by Gandin.⁹⁾ The value of A for Al–5mass%Si alloy was, firstly fixing $n=2.7$ for convenience, determined through fitting with the v_{tip} vs. ΔT profile in Fig. 5 of Ref. 1). As shown in Fig. 2, v_{tip} increases with the increase of local undercooling, ΔT , and the decrease of initial solute concentration, C^0 .

To simplify the entrapping judgment and the truncation, a coordinate transformation from xoy system to $x'C_vy'$ system is done for a growing cell v . As shown in Fig. 1, in $x'C_vy'$ system, the virtual growth center, C_v , of cell v is the origin, and the normal direction of growth plane $\langle 11 \rangle$ for cell v is x' -axis. When the coordinates of its neighbor cell μ in $x'C_vy'$ system is inside the square envelope with growth length, L_v^t (i.e. $|x'_\mu| \leq L_v^t$ and $|y'_\mu| \leq L_v^t$), it is entrapped. At that time, it is a random procedure for cell μ to be chosen to change state and grow up. This reflects the random growth mechanism in the actual solidification processes. Once the cell μ is chosen, it changes its state from liquid into solid and inherits the crystallographic orientation from its parent cell v . Its growth length, L_μ^t , is firstly inherited from L_v^t and then truncated to avoid overgrowth. For example, in the case shown in Fig. 1, through fixing the corner “A” which

Fig. 2. Growth kinetics of dendrite tip, as calculated by using the approximate expression which fits well with the predictions of the KGT model over a large range of undercoolings, for Al–Si alloys with three compositions. $v_{\text{tip}} = 1.7 \times 10^{-5} \times (\Delta T)^{2.7}$ m/s for Al–3mass%Si, $v_{\text{tip}} = 5.75 \times 10^{-6} \times (\Delta T)^{2.7}$ m/s for Al–5mass%Si, $v_{\text{tip}} = 2.9 \times 10^{-6} \times (\Delta T)^{2.7}$ m/s for Al–7mass%Si.

belongs to the growth envelope of cell v and is the nearest corner to cell μ , the growth length L_μ^t is obtained by truncating lengths of $L_{\mu[11]}^t$ and $L_{\mu[\bar{1}\bar{1}]}^t$ according to the diagonal of a CA cell, $\sqrt{2}l_{CA}$.

$$L_\mu^t = [\text{Min}(L_{\mu[11]}^t, \sqrt{2}l_{CA}) + \text{Min}(L_{\mu[\bar{1}\bar{1}]}^t, \sqrt{2}l_{CA})] / 2 \dots\dots(12)$$

$L_{\mu[11]}^t$ and $L_{\mu[\bar{1}\bar{1}]}^t$ are the distance from μ to the opposite plane $\langle 11 \rangle$ and $\langle \bar{1}\bar{1} \rangle$ of the growth envelope of cell v .

The coordinates of growth center C_μ are firstly calculated in $x'C_vy'$ system based on the topological relations of L_v^t and L_μ^t , and then they are re-transformed to the original xoy system. When all of its neighbors become solid, the cell v will stop growing.

3. Simulation Procedure

The development of solidification microstructure in Al–Si alloy ingot with cross section size $0.03 \text{ m} \times 0.03 \text{ m}$ is modeled using the CA-FD coupling model. The physical properties of Al–Si alloy are shown in Table 1. The corresponding values for Al–7mass%Si alloy are almost the same as those adopted by Feller and Beckermann.¹⁶⁾ As shown in Table 1, besides the dependence of thermal conductivity on temperature (K), both partition coefficient and latent heat of solidification are regarded as a function of Si concentration in the present work. Two operation parameters, C^0 , and $\dot{T}$, and six nucleation parameters, ΔT_V , $\Delta T_{V,\sigma}$, n_V^* , ΔT_S , $\Delta T_{S,\sigma}$, n_S^* , are chosen in the calculation. Initial Si concentration, C^0 , is set as 3 mass% Si, 5 mass% Si and 7 mass% Si, cooling rate in intermediate position, $\dot{T}$, is set as 0.7 K/s, 2.3 K/s and 5.0 K/s, respectively. Four nucleation parameters are fixed as $\Delta T_{V,\sigma} = 0.1$ K, $\Delta T_S = 1$ K, $\Delta T_{S,\sigma} = 0.1$ K, $n_S^* = 1/\lambda_1$, however, n_V^* , ΔT_V for heterogeneous nucleation in the bulk of liquid are changed in calculation cases, as listed in Table 2. Heat transfer coefficient, h , corresponding to each value of $\dot{T}$ for different C^0 is obtained beforehand through several attempts.

The numerical solution of CA-FD coupling model is performed in the following sequence:

- (1) Give operation parameters: C^0 , h , $\Delta T_{\text{superheat}}$, nucleation parameters: ΔT_V , $\Delta T_{V,\sigma}$, n_V^* , ΔT_S , $\Delta T_{S,\sigma}$, n_S^* , physical properties: C_p , κ , ρ , and time step: Δt .
- (2) Calculate enthalpy, H_{n+1} , from Eq. (2) with heat transfer boundary conditions.
- (3) Calculate concentration in liquid, C_{n+1} , from Eq. (6) with a guess of solid fraction $f_{S,n+1}$.
- (4) Calculate temperature, T_{n+1} , from Eq. (5).
- (5) Obtain accurate temperature, T_{n+1} , concentration, C_{n+1} , and solid fraction, $f_{S,n+1}$, through iterations of Eqs. (4), (5) and (6).
- (6) Interpolate the temperature field from FD mesh nodes into CA cells. Calculate local undercooling for each

Table 2. Nucleation parameters required in CA model for Al–Si alloy.

Max nucleation undercooling in the bulk, ΔT_V , K	Standard deviation in the bulk, $\Delta T_{V,\sigma}$, K	Nucleation density in the bulk, n_V^* , m^{-2}	Max nucleation undercooling at surface, ΔT_S , K	Standard deviation at the surface, $\Delta T_{S,\sigma}$, K	Nucleation density at the surface, n_S^* , m^{-1}
Parameter fitting	0.1	Parameter fitting	1	0.1	$n_S^* = 1/\lambda_1$

Fig. 3. Calculated cooling curves for Al-7mass%Si and Al-3mass%Si ingots with different cooling rate in intermediate position $\dot{T}$.

CA cell.

- (7) Calculate the nucleation probability by the increase of local undercooling in two CA time step, ΔT_n , ΔT_{n+1} , from Eqs. (8) and (9).
- (8) Calculate the grain growth by “decentred square” growth algorithm from Eqs. (10), (11) and (12).
- (9) Return to step (2) until the solidification is completed in the entire domain ($f_s=1$ for all FD nodes).

4. Results and Discussions

4.1. Cooling Curves

Figure 3 shows calculated cooling curves for Al-7mass%Si and Al-3mass%Si ingots with different cooling rates in intermediate position, $\dot{T}$. Heat transfer coefficients, h corresponding to these cases are also shown.

Generally, the slope of the curve becomes smaller temporarily due to the appearance of primary phase and eutectic phase. After the completion of solidification, the slope returns to the original state. Here the cooling rates at three positions (intermediate, center and surface of ingot) are defined by the ratio of temperature decrease to solidification time. The solidification time decreases with the increase of Si concentration, C^0 , with the increase of cooling rate in intermediate position, $\dot{T}$, and with the position closer to ingot surface due to the efficiency of heat transfer.

4.2. Morphologies of Solidification Microstructure

The grain growth is stopped once the dendrites of primary phase collide with each other. The collision point is recognized as the so-called “grain boundary”. The linked grains whose orientation difference is greater than 1.8 deg. are regarded as two distinguished grains.

4.2.1. Effects of Parameters on Solidification Morphologies

Figure 4 shows the final solidification microstructures of

Al-7mass%Si, Al-5mass%Si and Al-3mass%Si, as calculated using the CA-FD coupling model. The parameter effects on the size of columnar and equiaxed grains are examined carefully. Firstly, the effects of operation parameters (C^0 and $\dot{T}$) and nucleation parameters (ΔT_v and n_v^*) on the size (length and width) of columnar grains can be recognized. The length of columnar grains increases clearly with the increase of nucleation undercooling, ΔT_v , with the decrease of Si concentration, C^0 , and with the decrease of cooling rate in intermediate position, $\dot{T}$. This tendency comes from the dendrite tip growth kinetics which leads to the average length of columnar grain. Briefly, the average length of columnar grain is directly proportional to the growth rate of dendrite tip, $v_{tip}(\Delta T, C^0)$, and inversely proportional to $\dot{T}$.¹⁾ In addition, due to the implicit relationship between nucleation density, $n_v^* (=1/\lambda_1)$, and cooling rate at ingot surface, $\dot{T}_{Surface}$: $\lambda_1 = 220 \times 10^{-6} \times (\dot{T}_{Surface})^{-0.55}$, the width of columnar grain increases clearly with the decrease of $\dot{T}$. Here, it is noted that $n_v^* \geq 1/\lambda_1$, i.e. nucleation density at ingot surface is usually greater than columnar grain density ($1/\lambda_1$) due to the competition of the nucleation and the grain growth.

Secondly, the effects of operation parameters (C^0 , $\dot{T}$) and nucleation parameters (ΔT_v , n_v^*) on the size of equiaxed grains can also be recognized from Fig. 4. The size of equiaxed grains decreases clearly with the increase of nucleation density, n_v^* ; while it decreases somewhat with the increase of nucleation undercooling, ΔT_v , only for Al-3mass%Si alloy, as shown in Fig. 5. This tendency comes from the competition of the nucleation and the grain growth. Consequently, only nucleation density in the bulk liquid will control the size of equiaxed grains in the case of comparatively low dendrite tip growth rate. Here, the equiaxed grain size was evaluated by the grain size analysis software WINROOF. The size of each grain was determined as the area-equivalent diameter.

Fig. 4.

Fig. 4. The final solidification microstructures of Al-7mass%Si, Al-5mass%Si and Al-3mass%Si alloy ingots, as calculated by the CA-FD coupling model.

Fig. 5. Effects of operation parameters (C^0 , $\dot{T}$) and nucleation parameters (ΔT_v , n_v^*) on the size of the equiaxed grains, as calculated by the CA-FD coupling model.

4.2.2. Identification of Nucleant and Estimation of Nucleation Density in Actual Al-Si Alloy

The effects of impurities on the grain size in actual Al-Si alloy are discussed in the present work by using the CA-FD coupling model, because it is found that nucleation density in the bulk liquid, n_v^* mainly control the size of the equiaxed grains in the case of $\Delta T_v \leq 3 \text{ K}$ for Al-Si alloy.

Figure 6 shows the effects of nucleation density, n_v^* , nucleation undercooling, ΔT_v , and Si concentration, C_0 , on

Fig. 6. Effects of nucleation density n_v^* , nucleation undercooling ΔT_v and Si concentration C^0 on the grain size under cooling rate $\dot{T} = 0.7 \text{ K/s}$, as calculated by using the CA-FD coupling model, and the experimental data of grain size for actual Al-Si alloys.

grain size under cooling rate in intermediate position $\dot{T} = 0.7 \text{ K/s}$, as calculated using the CA-FD coupling model, and the experimental data of grain size in actual Al-Si alloy by Hutt *et al.*¹⁵⁾ ($\dot{T} = 1.0 \text{ K/s}$), Lee *et al.*¹⁰⁾ ($\dot{T} = 0.7 \text{ K/s}$) and Hoefs *et al.*¹⁷⁾ ($\dot{T} = 0.7 \text{ K/s}$). The experimental data of grain size is in the range of 600 to 700 μm for Al-3mass%Si alloy, 750 to 900 μm for Al-5mass%Si alloy and 900 to 1200 μm for Al-7mass%Si alloy. Regarding nucleation undercoolings, Hutt *et al.*¹⁵⁾ reported $\Delta T_v = 1.2 \text{ K}$ for Al-3mass%Si alloy, $\Delta T_v = 2.05 \text{ K}$ for Al-5mass%Si alloy and $\Delta T_v = 2.3 \text{ K}$ for Al-7mass%Si alloy. Murty¹⁸⁾ also reported $\Delta T_v = 1.9 \text{ K}$ for Al-7mass%Si alloy. Those results indicate that with the increase of impurity concentration caused by the increase of Al concentration in actual Al-Si alloy, the heterogeneous nucleation occurs easier and then ΔT_v decreases.

Firstly, from the agreement between the calculated average equiaxed grain sizes and the experimental ones in Fig. 6, the nucleation densities in Hutt's, Lee's and Hoefs' experiments are roughly evaluated as $n_v^* = 200$ to 300 cm^{-2} ($n_v = 2050$ to 3760 cm^{-3}) for Al-3mass%Si; $n_v^* = 100$ to 200 cm^{-2} ($n_v = 720$ to 2050 cm^{-3}) for Al-5mass%Si and $n_v^* = 50$ to 150 cm^{-2} ($n_v = 260$ to 1330 cm^{-3}) for Al-7mass%Si. Secondly, it is found that the impurity element Fe in the form Al_3Fe might act as a nucleant for heterogeneous nucleation, because actual Al-Si alloy is prepared using commercial purity Al (Al: 99.7 mass%, Fe: 0.16 mass%, Si: 0.11 mass%)¹⁹⁾ and consequently there is

Fig. 7. Relationship among Si concentration C^0 , Fe concentration and nucleation density n_v^* in actual Al–Si alloy prepared by using commercial purity Al.

0.1552 mass% Fe for Al–3mass%Si, 0.152 mass% Fe for Al–5mass%Si and 0.1488 mass% Fe for Al–7mass%Si, respectively. Actually, the grain size of Al–7mass%Si alloy is in the range of 300 to 400 μm with 0.1 mass% addition of Fe (*i.e.* total 0.2488 mass% Fe).^{18,19)} It corresponds to $n_v^*=600\text{ cm}^{-2}$ ($n_v=10\,630\text{ cm}^{-3}$) as indicated in Fig. 6(c). Relationship among Si concentration, C^0 , Fe concentration and nucleation density, n_v^* , in actual Al–Si alloy prepared by using commercial purity Al is summarized in Fig. 7.

4.3. Criteria for Columnar to Equiaxed Transition (CET)

Here, the comparison is made for Al–Si alloy between the CET criteria based on the solidification path predicted by the CA-FD coupling model and one based on the critical temperature gradient conditions predicted by Hunt's model and Stefanescu's model.

4.3.1. CET Criteria from Solidification Microstructures by CA-FD Coupling Model

The CET is controlled by the competition of the nucleation and the grain growth process, as described in Sec. 4.2.1. Consequently, three steady-state growth conditions occur: (1) fully columnar growth (fully C), (2) mixed columnar/equiaxed growth (C+E) and (3) fully equiaxed growth (fully E).

(1) CET for Fully C

From the solidification microstructures shown in Fig. 4, it is found that the influence of nucleation density, n_v^* , on the transition from fully C to C+E is unclear in the range of $n_v^*=30$ to 300 cm^{-2} , while the transition from fully C to C+E occurs easily with the decrease of nucleation undercooling, ΔT_v , with the increase of Si concentration, C^0 , and with the increase of cooling rate in intermediate position, $\dot{T}$. Thus, it indicates that the transition from fully C to C+E is controlled almost uniquely by the growth rate of dendrite tip. Figure 8 shows the dependence of the CET criteria (transition from fully C to C+E) on ΔT_v , C^0 and $\dot{T}$ based on the solidification microstructures in Fig. 4.

(2) CET for Fully E

From the solidification microstructures shown in Fig. 4, it is found that the influence of nucleation density, n_v^* , on the transition from C+E to fully E is clear in the range of $n_v^*=30$ to 100 cm^{-2} , but unclear in the range of $n_v^*=150$ to 300 cm^{-2} ; and the transition from C+E to fully E occurs

Fig. 8. Dependence of the CET criteria on ΔT_v , C^0 and $\dot{T}$ for fully columnar growth (fully C) and for fully equiaxed growth (fully E), based on the solidification microstructure predicted by the CA-FD coupling model.

easily with the decrease of nucleation undercooling, ΔT_v , with the increase of Si concentration, C^0 . In addition, the influence of cooling rate in intermediate position, $\dot{T}$, on transition from C+E to fully E was unclear. Thus, it indicates that the transition from C+E to fully E is controlled by both the nucleation and the growth rate of the dendrite tip. Figure 8 also shows the dependence of the CET criteria (transition from C+E to fully E) on ΔT_v , C^0 and $\dot{T}$ based on the solidification microstructures in Fig. 4. The range of ΔT_v corresponding to transition from C+E to fully E agrees well with that by Martorano and Gandin,¹⁴⁾ which is 2.6 K for Al–3mass%Si and 4.7 K for Al–7mass%Si, as shown in Fig. 8.

4.3.2. CET Criteria by Hunt's Model

In Hunt's model,²⁰⁾ the critical temperature gradient conditions for fully equiaxed (fully E) and for fully columnar growth (fully C) are approximately given by

$$G < G_{\text{critical}\pm\text{fully E}} = 0.617n_v^{1/3} \left\{ 1 - \frac{(\Delta T_v)^3}{(\Delta T_C)^3} \right\} \Delta T_C \quad \text{for fully equiaxed growth} \dots\dots\dots(13)$$

$$G > G_{\text{critical}\pm\text{fully C}} = 0.617(100n_v)^{1/3} \left\{ 1 - \frac{(\Delta T_v)^3}{(\Delta T_C)^3} \right\} \Delta T_C \quad \text{for fully columnar growth} \dots\dots\dots(14)$$

where G is the temperature gradient in solid/liquid (S/L) interface expressed in forward or backward difference approximation at the interface (see Fig. 9). n_v is the nucleation densities for three-dimensional geometry. ΔT_C is the local undercooling ahead of columnar dendrite front,²¹⁾

$$\Delta T_C = 2 \left\{ - \frac{2m(1-k^{S/L})C_0\bar{v}_L\Gamma}{D_L} \right\}^{1/2} = \left\{ \frac{\bar{v}_L C_0}{A} \right\}^{1/2} \dots\dots(15)$$

where $\bar{v}_L$ is the solid/liquid (S/L) interface velocity. It is approximately set as the moving velocity of liquidus isotherm (see Fig. 10). $A = D_L / (4 \cdot (-2)m(1-k^{S/L})\Gamma)$ [m mass% $\text{s}^{-1} \text{K}^{-2}$] is a constant, and physical properties are listed in Table 1.

The dependence of the CET criteria on nucleation undercooling, ΔT_v , and Si concentration, C^0 , is discussed under nucleation density, $n_v^*=150\text{ cm}^{-2}$ and cooling rate in inter-

Fig. 9. Distributions of temperature gradient at solid/liquid interface. At time t_i when position x_i becomes solid, temperature gradient is evaluated by the sequent temperatures of the closest positions forward and backward: $G_L = G_{\text{forward},i} = (T_{i+2}(t_i) - T_{i+1}(t_i)) / (x_{i+2} - x_{i+1})$, $G_S = G_{\text{back},i} = (T_i(t_i) - T_{i-1}(t_i)) / (x_i - x_{i-1})$.

Fig. 10. Moving velocity of solid/liquid interface. It is set as the moving velocity of the liquidus isotherm, that is, the ratio of the distance between two consecutive positions where the liquidus isotherm travels to the time elapsed, $\bar{v}_L = (x_{i+1} - x_i) / (t_{i+1} - t_i)$. It is drawn at the middle of the two adjacent positions, $(x_i + x_{i+1}) / 2$.

mediate position, $\dot{T} = 0.7 \text{ K/s}$. Comparison of the CET criteria based on the solidification path predicted by the CA-FD coupling model and the critical temperature gradient conditions predicted by Hunt's model for Al-7mass%Si alloy, Al-5mass%Si and Al-3mass%Si alloy are shown in **Fig. 11** to **Fig. 13**.

The symbols denoted by Δ , $\blacksquare$ and $\bullet$ on the solidification path by the CA-FD coupling model represent the fully columnar growth (fully C), the fully equiaxed growth (fully E) and the mixed columnar and equiaxed growth (C+E), respectively. As illustrated, the CET criteria based on the solidification path by CA-FD coupling model are strongly dependent on both ΔT_V and C^0 . The curves of the critical temperature gradient conditions by Hunt's model (the curves labeled $G_{\text{fully E}}$ and $G_{\text{fully C}}$) are also strongly dependent on both ΔT_V and C^0 . To well illustrate the CET, the solidification microstructures at typical stages of solidification are also given below the CET map.

As shown in Fig. 11 to Fig. 13, the good agreement is obtained between these two. Hunt's assumptions based on steady state seem to hold approximately, because the temperature field is changed slowly in the case of cooling rate $\dot{T} = 0.7 \text{ K/s}$.

4.3.3. CET Criteria by Stefanescu's Model

Stefanescu²¹⁾ also proposed the critical temperature gradient conditions for CET as follows,

$$G \leq G_{\text{CET}} = 3.22 \left[\frac{n_V}{f_{\text{S,coh}}} \left(1 - \frac{v_L}{\bar{v}_L} \right) \right]^{1/3} \frac{\mu_e}{\mu_c} \Delta T \dots (16)$$

Fig. 11. Comparisons between the CET criteria based on the solidification path predicted by the CA-FD coupling model and one based on the critical temperature gradient conditions predicted by Hunt's model and Stefanescu's model for Al-7mass%Si alloy. Upper part: dependence of the CET criteria on ΔT_V and C^0 under $n_V^* = 150 \text{ cm}^{-2}$ and $\dot{T} = 0.7 \text{ K/s}$. Lower part: solidification microstructures at typical stages of solidification.

Fig. 12. Comparisons between the CET criteria based on the solidification path predicted by the CA-FD coupling model and one based on the critical temperature gradient conditions predicted by Hunt's model and Stefanescu's model for Al-5mass%Si alloy. Upper part: dependence of the CET criteria on ΔT_V and C^0 under $n_V^* = 150 \text{ cm}^{-2}$ and $\dot{T} = 0.7 \text{ K/s}$. Lower part: solidification microstructures at typical stages of solidification.

Fig. 13. Comparisons between the CET criteria based on the solidification path predicted by the CA-FD coupling model and one based on the critical temperature gradient conditions predicted by Hunt's model and Stefanescu's model for Al-3mass%Si alloy. Upper part: dependence of the CET criteria on ΔT_V and C^0 under $n_V^* = 150 \text{ cm}^{-2}$ and $\dot{T} = 0.7 \text{ K/s}$. Lower part: solidification microstructures at typical stages of solidification.

where $f_{S,coh}$, the coherency solid fraction, is the solid fraction when dendrite coherency is established during columnar grain growth process. A reasonable limit is assuming that $f_{S,critical} = f_{S,coh}$ with $f_{S,critical}$ the critical solid fraction for CET. Stefanescu recommended a typical value of $f_{S,coh} = 0.2$ to 0.4. In the present work, $f_{S,coh} = 0.3$. v_L is the flow velocity by convection (here $v_L = 0$). μ_e and μ_c are the growth coefficient of equiaxed and columnar dendrites, respectively. In steady-state columnar growth, $\mu_c = (k^{S/L} D_L) / \{ \pi^2 \Gamma m (k^{S/L} - 1) C_0 \}$ and $\mu_e = 0.5 \mu_c$, when the thermal undercooling is ignored. The local undercooling in steady-state columnar growth can be deduced as $\Delta T = \sqrt{\bar{v}_L / \mu_c}$ from the expression of solid/liquid (S/L) interface velocity $\bar{v}_L = \mu_c \Delta T^2$. Stefanescu's expression was deduced without the assumption of steady-state columnar growth, although it includes the original idea of coherency solid fraction and liquid convection.

The critical temperature gradient conditions by Stefanescu's model for Al-7mass%Si alloy, Al-5mass%Si and Al-3mass%Si alloys are added in Fig. 11 to Fig. 13. The curves of the critical temperature gradient conditions by Stefanescu's model (the curves labeled G_{CET}) are dependent on C^0 , but independent on ΔT_V , because the term of ΔT_V is not included in Stefanescu's expression.

5. Conclusions

A CA-FD coupling model has been developed to analyze the development of solidification microstructure and the CET criteria for Al-Si alloy. The validation of the model was achieved with the help of previous experiments. The

following conclusions can be obtained:

(1) Through the examination of the effects of operation parameters and nucleation parameters on solidification morphologies, the length of columnar grains is controlled by the dendrite tip growth kinetics, and the width of columnar grains is controlled by the implicit relationship between nucleation density and cooling rate at ingot surface. The size of equiaxed grains is controlled by the competition of the nucleation and the grain growth.

(2) With the controllability of nucleation density in the bulk of liquid for equiaxed grain size, the nucleant and the nucleation density in actual Al-Si alloy are estimated. The impurities in actual Al-Si alloy prepared by using commercial purity Al, especially the element Fe in the form of Al_3Fe , might act as a heterogeneous nucleant and do contribute to the grain refinement.

(3) The CET criteria based on both the solidification path predicted by the CA-FD coupling model and the curves of the critical temperature gradient conditions predicted by Hunt's model are strongly dependent on nucleation undercooling and Si concentration. A good agreement is obtained between these two.

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