A numerical study of dendrite growth and microstructure transition in a non-equilibrium solidification

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Abstract. Controlling the thermal condition of a solidification process to obtain a desirable microstructure morphology and then prepare specific materials with excellent mechanical properties or special functions has been attracting many concerns, where the behavior of microstructure morphology transition, especially the columnar-to-equiaxed transition, is one of the most important fundamental issues. This paper introduced a kinetic model for dendrite growth with a large undercooling during a non-equilibrium solidification process using cellular automaton, in which the local thermodynamic equilibrium condition, non-equilibrium partitioned solute, curvature and interface energy anisotropy were considered. The growth rate of dendrite tip was numerically investigated by using the cellular automaton model, and the effects of temperature gradient and cooling rate on the growth rate and morphology features were found. By integrating the simulation results and analytical models, a new microstructure morphology transition criterion for the non-equilibrium solidification depending on the temperature gradient and cooling rate was proposed, in which both the planar-to-columnar and the columnar-to-equiaxed transition were presented.

1. Introduction

Since the microstructure characteristics greatly affect the mechanical properties of solidified alloy materials, it is of great merit to understand the solidification process and take insight into the growth kinetics of dendrites. Columnar and equiaxed grains are two typical solidified morphology characteristics that greatly determine the quality of a cast product [1]. However, controlling the microstructure morphology transition as expected is difficult, especially in a non-equilibrium solidification process of metals and alloys with a high cooling rate and large temperature gradient, such as the selective laser melting [2-4] and arc welding [5] processes. It is a typical phenomenon in most non-equilibrium solidification processes that columnar dendrites derived from epitaxial growth appear first and then the columnar-to-equiaxed transition (CET) could follow up when the front of a columnar dendrite is blocked by the growing equiaxed grains in the constitutionally undercooled liquid ahead of the columnar grain [6-7]. Either refining the columnar grains or promoting the CET is an important way to modulate the mechanical properties of a solidified part, and many theoretical and experimental studies were devoted to regulating the microstructure by operating the process parameters [8].

Many efforts have been done to try to understand the quantitative effect of the temperature field and the temperature-related process parameters on the nucleation and growth of dendrites. Hunt [9] firstly gave a simple expression to predict the appearance of fully equiaxed grains during a directional solidification and provided a criterion for qualitatively discussing the equiaxed growth in more complicated casting situations. Gäumann [10] proposed a CET condition, that if the ratio of \( G/V \) (G:}
temperature gradient, $V$: dendrite growth rate) is lower than a system constant, for the solidification of multicomponent alloys under large temperature gradients. However, the dendrite growth rate, depending on many factors not only the temperature field, is difficult to be measured directly in practice. The solidified microstructure is commonly predicted by using temperature gradient $G$ and cooling rate $R$ at present, and with the decrease of $G/R$, the microstructure shows planar, cellular, columnar, and then equiaxed [11-12]. Unfortunately, this relationship has not been quantitatively presented. In recent years, numerical simulation methods have been used to examine the dependence of microstructure evolution on solidification conditions, particularly the cooling rate and temperature gradient [13-14], in which the cellular-automata is one of the widely used methods.

This work aims to present a new theoretical conception for the microstructure transition, especially the CET, during a non-equilibrium solidification process with a large undercooling by considering the local thermodynamic equilibrium condition and the non-equilibrium partitioned solute at the S/L interface. Combining the numerical study with the analytical formulas, we will propose a new CET criterion that can be evaluated explicitly from the temperature gradient, cooling rate, nucleation rate instead of the growth rate of dendrite tips. This CET criterion builds a bridge between the microstructure transition and process parameters in practice. Taking the 718 alloys as an example, this work will present a comprehensive understanding of the microstructure transition under a large undercooling and compose the microstructure spectrogram under a wide thermal condition. We believe that this work can be helpful for the microstructure control during a non-equilibrium solidification process with a large undercooling, such as the metal additive manufacturing, selectively-laser-melting and arc-welding processes, etc.

2. **Theory and cellular-automata model**

For an alloy, the dendrite growth is generally considered to be mainly affected by temperature field and material chemical composition, that is, thermodynamic undercooling and constitutional undercooling in the equilibrium phase diagram. When the temperature is lower than the liquidus temperature of the alloy liquid, the dendrites begin to nucleate and grow, and the solute is rejected from the solid phase into the liquid phase at the front of the S/L interface, resulting in a decrease in the liquidus temperature, as a consequence, further growth of the S/L interface requires a lower temperature or a lower solute concentration at its front. The S/L interface of a metal crystal is of nonfaceted structure, the required kinetic undercooling for the growth is fairly small, so the S/L interface can be considered in a kinetic equilibrium state at each time step. Additionally, both the S/L interface curvature and interface energy anisotropy are also important to the dendrite growth, which can be ignored during a near-equilibrium solidification but must be considered in a large undercooling. Hence, the formula for the kinetic equilibrium in the liquid at the front of the S/L interface can be given by [15]:

$$\Delta T = m(C_0 - C_i^*) + \Gamma \kappa f(\hat{n}),$$

(1)

where $\Delta T$ is the undercooling at the S/L interface. $m, C_0,$ and $C_i^*$ are the liquidus slope, initial concentration of the original alloy, and the solute concentrations in the liquid phase at the front of the S/L interface, respectively. $\Gamma$ is the Gibbs-Thomson coefficient, $\kappa$ the mean curvature, given by [16]:

$$\kappa = \frac{1}{d}(1 - \frac{2}{N+1} \sum f_{si}),$$

(2)

where $f_{si}$ represents the solid fraction of a cell and its nearest neighbor cells, $d$ the cell size, $N=27$ for a three-dimensional case. The anisotropy function $f(\hat{n})$ is defined as [17]:

$$f(\hat{n}) = \gamma_0 (1 - 3\varepsilon)[1 + \frac{4\varepsilon}{1 - 3\varepsilon} (\cos^4 \phi + \sin^4 \phi(1 - 2\sin^2 \theta \cos^2 \theta))],$$

(3)

where $\gamma_0$ is the isotropic interfacial energy, $\varepsilon$ is the strength of the fourfold anisotropy, $\phi$ and $\theta$ are the two standard spherical angles with the interface normal direction, given by:
\[ \theta = \arccos \left( \frac{\partial f_x}{\partial x} \sqrt{\left( \frac{\partial f_x}{\partial x} \right)^2 + \left( \frac{\partial f_y}{\partial y} \right)^2} \right), \] (4)

\[ \varphi = \arccos \left( \frac{\partial f_z}{\partial z} \sqrt{\left( \frac{\partial f_x}{\partial x} \right)^2 + \left( \frac{\partial f_y}{\partial y} \right)^2 + \left( \frac{\partial f_z}{\partial z} \right)^2} \right). \] (5)

The relationship between the solute concentration in the liquid phase \( C_l^* \) and the solid phase \( C_s^* \) in the immediate vicinity of the S/L interface can be expressed as \([18]\):

\[ C_l^* = k_e C_s^* = \frac{k_0}{k_0 + (1-k_0) \exp(-aV/D_l)} C_s^*, \] (6)

where \( k_e \) and \( k_0 \) are the solute partition coefficients in the non-equilibrium and equilibrium states, respectively, \( a \) is the atomic spacing, \( V \) the solidification velocity, \( D_l \) the solute diffusivity in the liquid.

To maintain the kinetic equilibrium always in the liquid phase at the front of the S/L interface, once the actual concentration \( C \) is lower than the equilibrium concentration \( C_l^* \), a certain amount of solute will be rejected by the increasing solid phase, reaching a new kinetic equilibrium state. The increment of the solid fraction at each time step can be computed by \([10]\):

\[ \Delta f = \left( C_l^* - C \right) \left[ C_s^* (1-k_e) \right]. \] (7)

The solidification time is very short during a rapid solidification process, indicating that the diffusion only occurs in a very thin layer at the S/L interface, as a consequence, convection effect is slight and can be ignored. Thus the governing equation of the concentration field without convection term can be written as:

\[ \frac{\partial C}{\partial t} = \nabla \cdot D \nabla C + C \left( 1-k_e \right) \frac{\partial f}{\partial t}, \] (8)

where \( D \) is the solute diffusion coefficient. The last term on the right-hand side is a concentration source term due to the solute rejection.

Here, we apply a cellular automata method (please see our previous work \([19]\) for details) to model the dendrite growth, while the way to computing the concentration is improved to ensure concentration conservation during the numerical computation of the model.

3. Numerical study of dendrite growth

The morphology of equiaxed and columnar crystals, the growth rate of dendrite tips, and the primary dendrite arm spacing of columnar dendrites determined by the undercooling are the important issues for the microstructure transition. Additionally, the driving force for the growth of a solid-liquid interface comes from the combined effect of temperature, concentration, and interface curvature. However, the concentration and interface curvature greatly depend on the temperature. Consequently, we first carry out a series of three-dimensional simulations to find the quantitative relationships between the dendrite growth and the undercooling. These simulations are computed on a grid resolution of 0.1×0.1×0.1 \( \mu \)m and using a fixed time step of 0.1 \( \mu \)s, and the used parameters and material properties are listed in Table 1.

The growth rate of dendrite tips is a critical quantity to calculate the primary dendrite arm spacing, the CET criterion as well as the microstructure evolution. By taking the derivation of the displacement to the growth time, we obtain the growth rate quantitatively. The averaged growth rates under different undercoolings are plotted in Figure 1 and compared with Nastac’s prediction \([21]\). By fitting the data under a large undercooling (\( \Delta T > 5 \) K) using a quadratic function, we have the relationship between the growth rate of the dendrite tips and the undercooling as:

\[ V = \mu \Delta T^2 \text{ with } \mu = 3.2 \times 10^{-5} \text{ [ms}^{-1} \text{K}^{-2}]. \] (9)
undercooling. The difference mainly results from a constant solute partition coefficient considered in the Nastac’s prediction, however, the solute partition coefficient should be variable and greater in a non-equilibrium solidification with a large undercooling than that in the equilibrium, which results in a smaller $\mu$ and then a lower growth rate, see the Nastac’s model for details [21].

Table 1 Simulation parameters and material properties for Inconel 718 alloy.

| Property and symbol       | Value              | Ref.  |
|---------------------------|--------------------|-------|
| Liquidus temperature $T_l$ (K) | 1609               | [20]  |
| Solidus temperature $T_s$ (K) | 1533               | [20]  |
| Initial solute concentration $C_0$ (wt.%) | 5.0                | [1, 13] |
| Liquidus slope $m$ (K·wt.%$^{-1}$) | -10.5              | [1, 13] |
| anisotropy coefficient $\varepsilon$ | 0.03               | [13]  |
| Partition coefficient $k_0$ | 0.48               | [1, 13] |
| Gibbs-Thomson coefficient $\Gamma$ (K·m) | $3.65 \times 10^{-7}$ | [1, 13] |
| Liquid diffusion coefficient $D_L$ (m$^2$·s$^{-1}$) | $3 \times 10^{-9}$ | [1, 13] |

Fig.1 Comparison of the growth rate of the dendrite tip between the simulation and Nastac’s prediction.

4. Microstructure transition
To examine the microstructure transition of the Inconel 718 alloy, a series of cases with an initial crystal nucleus at the bottom of the domain were simulated, in which parameter sweeping of the temperature gradient and cooling rate was done. The spectrogram of the dendrite morphologies is shown in Figure 2. These morphologies present three significant categories: the planar region on the top, the columnar region in the middle, and the equiaxed region at the bottom. When the temperature gradient is high up to $1 \times 10^8$ K/m, the dendrites will grow into planar crystals, however, if the cooling rate is extremely high meanwhile, saying higher than $1 \times 10^6$ K/s, the stability of the flat solid-liquid interface could be destroyed and the planar crystal might turn into an amorphous structure. If the temperature gradient is reduced, the dendrite tips will turn into a columnar structure and the columnar crystal region gradually expands as the cooling rate increases. If the temperature gradient continues to be reduced to the range of $1 \times 10^6$ K/s - $1 \times 10^7$ K/s, the CET behavior will occur. Meanwhile, it can be seen that both reducing temperature gradient and increasing subcooling can promote CET. If the temperature gradient is reduced to less than $1 \times 10^6$ K/m, the crystal nucleus usually grows up to be an equiaxed shape with a large arm along the $Z$ direction and four slightly shorter dendritic arms on the bottom plane. If increasing the cooling rate, the arm along the $Z$ direction significantly becomes
shorter and a small-sized equiaxed crystal is obtained, on which the secondary dendrite arms gradually also become shorter and even disappear.

To give a more practical quantitative criterion for the microstructure transition, which depends on the temperature gradient and cooling rate instead of the dendrite growth rate, we collect the relationships between the temperature gradient, cooling rate, dendrite growth rate and the undercooling and the aforementioned simulation results to determine the demarcation lines analytically for the spectrogram of the dendrite morphology. There is a well-known relationship between the undercooling and the local cooling rate along with a positive temperature gradient, given by:

$$\frac{d(\Delta T)}{dt} = R - \frac{GV}{\Delta T}.$$  \hspace{1cm} (10)

Assuming that the dendrites grow in a steady-state [9], we have $R=GV$. Hence, the compositional undercooling criterion $G/V < (T_f-T_s)/D_l$ determining the transition of a stable planar front to unstable columnar or equiaxed crystals can be rewritten as:
\[ \frac{G^2}{R} < \frac{T_i - T_c}{D_i}. \]  

(11)

Taking the Inconel 718 alloy as an example, a stable planar front could appear theoretically when \(G^2/R=2.53\times10^{10} \text{ K}\cdot\text{s/m}^2\). In practice, this condition is hard to be reached, and thus the planar crystal usually cannot be found during an actual solidification process.

The CET is common in an actual solidification, which can be properly controlled to obtain excellent mechanical properties (fully equiaxed crystals) or special functional materials (columnar crystals or single crystals). The CET is a phenomenon that a certain amount of crystal nuclei are produced at the tips of a dendrite in a supercooled liquid. These crystal nuclei will grow into equiaxed crystals, which could block the further growth of the columnar crystals by mechanical interaction [6] or solute interaction [22]. Our simulation results agree with the mechanical-interaction hypothesis more, in which the volume fraction of equiaxed grains is an important indicator if the growth of columnar dendrites is blocked. According to the Avrami equation [13], the volume fraction

\[ \phi = 1 - \exp \left( -\frac{4}{3} \pi r^3 N_0 \right), \]

which can be evaluated by the nucleation rate \(N_0\) and the grain radius \(r\).

The grain radius is given by:

\[ r = \int_0^\tau V dt = \frac{\mathcal{M}}{3R} (\Delta T^3 - \Delta T_n^3), \]

(12)

where \(\tau\) is the growth time of the nucleation undercooling varying from \(\Delta T_n\) to \(\Delta T\). Applying the relationship between the temperature gradient and the undercooling, \(G = N_0^{1/3} \Delta T\) proposed by [6], we have a new criterion describing the CET as:

\[ \frac{G^3}{R} = \frac{1}{9\mu} \left( \frac{4\pi N_0}{3\ln(1-\phi)} \right)^{2/3} \left( 1 - \frac{\Delta T_n^3}{\Delta T^3} \right)^2. \]

(13)

Defining a ratio \(\omega = \Delta T_n/\Delta T\), we then have the CET criterion for Inconel 718 alloy \((N_0=2.65\times10^{14} \text{ m}^3\) [21]) that the microstructure morphology is of fully equiaxed grains when \(G^3/R<4.85\times10^{13}(1-\omega^3)\) K^2\cdot\text{s/m}^3, fully columnar grains when \(G^3/R>1.06\times10^{15}(1-\omega^3)\) K^2\cdot\text{s/m}^3, and mixed columnar/equiaxed grains otherwise.

In summary, the morphology spectrogram of the Inconel 718 alloy with quantitative demarcation lines at different undercooling coefficients is shown in Figure 3. This spectrogram is split by the demarcation lines into four regions: I: planar, II: full columnar, III: mixed of columnar and equiaxed, and IV: full equiaxed. It can be seen that as the temperature gradient decreases and the cooling rate increases, the dendrite morphology will experience I to IV, and the temperature gradient shows a more significant impact on the morphology transition while the cooling rate greater on the dendrite size. In practice, it is difficult to increase one and decrease the other simultaneously to obtain equiaxed crystals due to the temperature gradient and the cooling rate are usually positively correlated. Generally, to obtain fine equiaxed crystals, rapid cooling and an alloy with a large heat transfer coefficient are required [23, 24]. On the contrary, to obtain columnar crystals, which is usually desired in directional solidification, the alloy with a small heat transfer coefficient is preferred. Moreover, Figure 3 also shows the effect of the ratio \(\omega\) on the demarcation lines. The results indicate that the demarcation lines are almost unchanged when the ratio is less than 0.8 while the lines between II and III as well as III and IV move towards the bottom right visibly when the ratio increases near to 1, resulting in the full-columnar region expanded while the full-equiaxed region narrowed rapidly. Hence, reducing the ratio \(\omega\) is a more practical way to obtain a fine equiaxed crystal structure, which can be achieved by adding grain refiners to increase the heterogeneous nucleation rate. Additionally, for the columnar crystal with strong anisotropy preferred in preparing some functional materials and single-crystal materials, it should increase the ratio \(\omega\) to expand the columnar-crystal region by reducing the nucleation undercooling.
5. Conclusion
This work developed a kinetic model of dendrite growth based on a cellular-automata method, which includes the local thermodynamic equilibrium condition, non-equilibrium partitioned solute, curvature and interface energy anisotropy. This model was applied to present the fundamental characteristics of the morphology and growth of the dendrite as well as the secondary arms, and the effects of temperature gradient and cooling rate on them were found. A quantitative relationship between the growth rate of dendrite tips and the undercooling was obtained. A new microstructure transition criterion depending on the temperature gradient, cooling rate and nucleation undercooling were given, especially for the CET criterion, and the spectrogram of the dendrite morphology of the Inconel 718 were composed. The contribution of this work is more straightforward to guide the morphology control of the Inconel 718 by manipulating the thermal condition in the solidification with a large undercooling.

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