Simulation of microstructural evolution in directional solidification of Ti-45at.\%Al alloy using cellular automaton method

*Wang Kuangfei\(^1\)\(^,\) Lu Shan\(^2\), Mi Guo\(^a\)\(^1\), Li Changyun\(^a\)\(^1\) and FU Hengzh\(i\)\(^1\)\(^,\)\(^2\)

(1. School of Materials Science and Engineering, Henan Polytechnic University, Jiaozuo, 454100, China; 2. School of Electromechanical Engineering, Jiaozuo University, Jiaozuo 454100, China)

Abstract: The microstructural evolution of Ti-45 at.\%Al alloy during directional solidification was simulated by applying a solute diffusion controlled solidification model. The obtained results have shown that under high thermal gradients the stable primary spacing can be adjusted via branching or competitive growth. For dendritic structures formed under a high thermal gradient, the secondary dendrite arms are developed not very well in many cases due to the branching mechanism under a constrained dendritic growth condition. Furthermore, it has been observed that, with increasing pulling velocity, there exists a cell/dendrite transition region consisting of cells and dendrites, which varies with the thermal gradient in a contradicting way, i.e. increase of the thermal gradient leading to the decrease of the range of the transition region. The simulations agree reasonably well with experiment results.

Key words: Ti-Al based alloy; directional solidification; solute diffusion controlled mode; cellular automaton

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During directional solidification of alloys, the cell/dendrite transition is one of the most fundamental and important solidification phenomena \[^{[1,2]}\], because their columnar grain spacing is an important structural parameter reflecting the solidification process. According to Mullins–Sekerka theory of morphological instability \[^{[3]}\], the growth of regular cells occurs in low growth rate (V) conditions. As the temperature gradient \((G_L)\) is reduced and the growth rate is increased, cells begin to branch, leading to dendrites. Theoretically, the critical velocity of these transitions can be determined, however due to the complexity and gradual evolution process, such as the high nonlinearity, detailed cell/dendrite evolution mechanism has yet to be well understood and further research is necessary.

Two significantly controllable variables, the temperature gradient \((G_L)\) and pulling velocity \((V_p)\), are independently controlled and held constant with time during directional solidification experiments. The evolution of simulated microstructures can be directly visualized and compared with actual microstructures from experiments. The present simulation was mainly to investigate the microstructural evolution of Ti-45at.\%Al alloy during directional solidification with initial concentration chosen in range of the \(L\rightarrow L+\beta(Ti)\) liquid–solid transition \[^{[4]}\] in the phase diagram. The calculated results were presented and discussed.

1 Model description

The model is based on the following assumptions: (1) The alloys are molten, quiescent and uniformly mixed, and have no melt convection; (2) The melt is only cooled at the bottom, and the top and side walls are insulated; (3) No diffusion occurs in the solid phase, and concentration distribution reaches local equilibrium at the solid/liquid interface; (4) No nucleation occurs in the bulk liquid, and only the growth of the seeds placed at the base of the directionally solidified ingot will be considered.

1.1 The model of grain growth

The structure of the stochastic model is similar to that described in ref. \[^{[5]}\]. It consists of a regular network of cells that resembles the geometry of interest. The model is characterized by: (a) geometry of the cell; (b) state of the cell; (c) neighborhood configuration and (d) several transition rules that determine the state of the cell. In this work, the geometry of the cell is a square. Each cell has three possible states: liquid \((f_s = 0)\), interface \(0 < f_s < 1\), or solid \((f_s = 1)\).

It is assumed that a nucleus formed at a particular location will grow based on the growth velocity of the \(S/L\) interface obtained from Equ. (2) and the neighborhood configuration rule previously described \[^{[5]}\]. As a "liquid" cell nucleated and became an active "interface" cell \((0 < f_s < 1)\), it would grow until fully solidified \((f_s = 1)\). Thereafter, the "interface" cell would capture the neighboring cells if a randomly generated number, rand, is smaller than the capture probability, \(p_c\), defined as follows \[^{[5]}\]:

\[
p_c = \frac{1}{\sqrt{\tan \theta} + 1}
\] (1)

\(^{*}\)Wang Kuangfei

Male, born in 1965, associate professor, Ph.D. He obtained his doctor’s degree from Harbin Institute of Technology, and his research interest focuses on the application of computer technology in casting.

E-Mail: wkf-12@hpu.edu.cn

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Where $\theta$ is the growth angle and takes values from $-\pi/4$ to $\pi/4$. The growth velocity of the S/L interface is calculated according to solute conservation at the S/L interface:

$$c'_i - c^* = \nabla \cdot (V_i \nabla c^*) = D_i \nabla^2 c^* \cdot \hat{n}$$

(2)

Where $V^*_i$ is the normal velocity of the interface, $c^*_i$ the solute concentration in liquid of the interface, $c'_i$ the solute concentration in solid of the interface, $D_i$ the liquid solute diffusion coefficient, $\hat{n}$ the interface normal vector, and $c$ the solute concentration in liquid ($c_L$) or solid ($c_s$) phase.

The interface liquid concentration $c^*_i$ is defined as:

$$c^*_i = c_0 + \frac{1}{m_i}[T_c - \Gamma \kappa f(\phi, \theta) - T(t)]$$

(3)

Where $T_c$ is the equilibrium liquidus temperature of the alloy, $m_i$ the liquidus slope in the phase diagram, $\kappa$ the mean curvature of the S/L interface, $\Gamma$ the Gibbs–Thomson coefficient, $f(\phi, \theta)$ is a coefficient used to account for growth anisotropy, and $\phi$ the preferential crystallographic orientation angle. $\kappa$ and $f(\phi, \theta)$ are described in Ref. [5], $T(t)$ is the local temperature, and $c_0$ is the initial concentration of the liquid.

Knowing the velocity components in both $x$- and $y$-directions, the solid fraction increment is calculated with:

$$\delta f_s = \frac{\delta t}{a}(V_x + V_y - V_y V_x \delta t)$$

(4)

Where $a$ is the mesh size (uniform and constant for both $x$- and $y$- direction), and $\delta t$ is the time step. Then, the solid fraction can be expressed as:

$$f_{s}^{p+1} = f_{s}^{p} + \delta f_s$$

(5)

Where superscripts "$p+1$" and "$p$" denote the new and the old solid fraction values, respectively.

### 1.2 The concentration fields in the liquid

Assuming the equilibrium partition coefficient, it is obtained that the partitioning of solute in the growing cell is determined by:

$$c'_s = k_0 c^*_i$$

(6)

Where $k_0$ is a constant partition coefficient. The diffusion equations in liquid and solid can be expressed in a similar form:

$$\frac{\partial c^*_i}{\partial t} = \nabla \cdot (D_i \nabla c^*_i) + c'_i (1 - k_0) \frac{\partial f_s}{\partial t}$$

(7)

$$\frac{\partial c^*_s}{\partial t} = \nabla \cdot (D_s \nabla c^*_s)$$

(8)

The time step used in calculations is given by:

$$\delta t = \frac{1}{5} \min\left(\frac{a}{V_{\text{max}}} \frac{a^2}{D_i}, \frac{a^2}{D_s}\right)$$

(9)

Where $a$ is the mesh size (uniform and constant for both $x$- and $y$-directions), $V_{\text{max}}$ the maximum growth velocity obtained by scanning the growth velocities ($V^*_i$) of all “interface” cells during each time step, and $D_i$ the solute diffusion coefficient in solid.

### 2 Results and discussion

In the present simulation of directional solidification, it is assumed that there is a constant positive liquid thermal gradient ($G(t)$), and that the local temperature $T(t)$ is given by

$$T(t) = T^* + G(t)(x - V_x t)$$

(10)

Where $V_x$ is the pulling velocity, $T^*$ is the reference temperature, and $t$ the local solidification time, and $x$ is the distance from the bottom of the directionally solidified domain.

In the following simulations, it is also assumed that no nucleation occurs in the bulk liquid; only the growth of the seeds placed at the bottom of the directionally solidified domain is considered. Therefore, the final microstructure develops only by branching and overgrowth mechanisms from these initial seeds. The undercooling available for the nucleation of the seeds was taken to be 1 K throughout this work. Several simulations were run for directional solidification from fixed numbers of seeds of 4 and the preferred growth direction of all the seeds <100> were perfectly aligned with the grid. Computational domain of 160x300 mesh was chosen with a cell edge length of 5 μm. A zero flux boundary condition was applied to the left and the right side of the domain, while keeping the liquid concentration at top of the domain constant, $c_0$. The material properties and the model parameters used in these simulations are given in Table 1.

### Table 1: Material properties and model parameters used in the simulation

<table>
<thead>
<tr>
<th>Properties</th>
<th>Variable</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Liquidus temperature</td>
<td>$T_L$</td>
<td>1,830 K</td>
</tr>
<tr>
<td>Liquidus slope</td>
<td>$m_L$</td>
<td>-8.9 K/at.%</td>
</tr>
<tr>
<td>Partition coefficient</td>
<td>$k_0$</td>
<td>0.63</td>
</tr>
<tr>
<td>Solute diffusion coefficient in liquid</td>
<td>$D_L$</td>
<td>$3.0 \times 10^{-8}$ m²/s</td>
</tr>
<tr>
<td>Solute diffusion coefficient in solid</td>
<td>$D_s$</td>
<td>$3.0 \times 10^{-12}$ m²/s</td>
</tr>
<tr>
<td>Gibbs-Thomson coefficient</td>
<td>$\Gamma$</td>
<td>$1.5 \times 10^{-7}$ K m⁻¹</td>
</tr>
<tr>
<td>Initial concentration</td>
<td>$c_0$</td>
<td>45at.%</td>
</tr>
</tbody>
</table>

A typical simulated columnar grain evolution for Ti-45at.%Al alloy is shown in Fig.1. The gray zones represent different solute constitutions in the solid and liquid zones. It is demonstrated that the dendritic stems directly grow from four seeds with a spacing of 200 μm, accompanied with some parallel secondary dendritic arms at both sides of the stems (Fig.1(a)). Further solidification led to the development of secondary dendrites perpendicular to the heat transfer direction and the formation of some tertiary dendrites emanating from the secondary dendrites and following along the direction of the primary dendrites. Apparently, they were all blocked by other secondary dendrites, as shown in Figs.1(b) and (c).

In addition to the branching of the dendrite arms, the simulation has also revealed phenomena that occurred during the columnar dendritic growth, such as selection and competition. It can be seen in Fig. 1(c) that branching competition must have occurred during the dendrite growth. When the primary dendrite spacing is larger, the secondary dendrites are well developed, which is similar to that reported in ref. [7] on Ti-44 at% Al alloy. This result suggests that the current model can not only simulate the dynamic growth of the columnar dendrites, but also maybe applied to the branching and competition growth processes.
2.1 The effect of thermal gradient

In directional solidification, the thermal gradient is an important factor affecting the shape of the solidification front. The effect of thermal gradient on structure evolution developed under the pulling velocity of 0.1 mm/s was simulated (shown in Fig. 2). At $G_t=5$ K/mm, dendrites form directly from all four seeds (Fig. 2(a)). Increasing the thermal gradient, the growth of side branches of dendrites is suppressed, and the primary and secondary spacing are shortened (Fig. 2(b)). At $G_t=20$ K/mm, cellular arms are developed from the bottom, and the primary spacing is significantly reduced to 50 μm (Fig. 2(c)). Finally, increasing thermal gradient to 40 K/mm, a superfine cellular interface is obtained in Fig. 2(d). It should be pointed out that superfine cellular morphology can be found under high thermal gradient conditions (Fig. 2(e)-(f)).
In order to characterize the dendrite morphology quantitatively, a series of simulations with the same initial nucleation conditions have been then carried out with pulling velocity ranging 0.25–1.0 mm/s, and at different temperature gradients. The effect of temperature gradient $G_L$ on the primary spacing is shown in Fig.3. It can be seen that at the same pulling velocity, increasing thermal gradient decreases the final primary dendrite spacing, and at the same thermal gradient, increasing pulling velocity also decreases the final primary dendrite spacing.

2.2 The effect of pulling velocity

Figure 4 shows the effect of the pulling velocity on the structural evolution where $G_L=10$ K/mm. At $V_p=0.001$ mm/s, a planar grain is developed from the seeds (Fig.4(a)). When pulling velocity increases to 0.005 mm/s, cells are formed directly from the seeds (Fig.4(b)-(c)). As $V_p \geq 0.1$ mm/s, some cells show a tendency to form secondary dendrite arms, and a transient structure composed of cells and dendrites is observed (Fig.4 (d)-(e)). When $V_p > 0.30$ mm/s, dendrites grow directly from the seeds, and as solidification proceeding, tertiary dendrites gradually emanate from the secondary dendrites, leading to the reduction of the primary spacing (Fig. 4(f)).

The comparison between experimental and modeling results is shown in Fig.5. With a low pulling velocity, at quenching, large quantity of parallel dendrites form at interface and in liquid phase, while fine cells appear opposite to the heat transfer direction (Fig.5(a)), which is close to the simulated results in Fig.5(b). It can be seen that, as the pulling velocity increases, coarse cells form during directional solidification (Fig.5(c)), which is also similar to the simulated results in Fig.5 (d).

Figure 6 illustrates the evolution of primary cell and dendrite arm spacing with the pulling velocity. Obviously, the columnar arm spacing is intensively dependent on pulling velocity and thermal gradient. With increasing pulling velocity, the microstructure can be classified into three regions: (1) cellular structure; (2) cell/dendrite transition; (3) regular dendrites. In the cellular structure region, the primary spacing increases with the pulling velocity. In the cell/dendrite
transition region, the range of the pulling velocity varies with the thermal gradient, but in general, the spacing increases with the decrease of thermal gradient.

In the regular dendrites region, the primary dendrite arm spacing decreases proportionally with the increasing pulling velocity. Similar transient structure was also experimentally observed in Ti46Al alloy [10], characterized by the increase of cell arm spacing with increasing growth rate. Through regression analysis, the variation of dendrite arm spacing, $\lambda_p$, with the pulling velocity and temperature gradient ($G_L=10$ K/mm) can be expressed as follows:

$$\lambda_p = K_1 G_L^{-0.5} V_p^{-0.24}$$  \hspace{1cm} (11)

Where $K_1$ is a material constant. The value of the material constant determined by linear regression analysis is $K_1=1.296 \times 10^{-3}$. The rate exponent $–0.24$ is close to a theoretical value of $–0.25$ resulting from the models of Kurz and Fisher [11].

$$\lambda_p = 4.3 \left( \frac{\Gamma D_1 \Delta T_k}{k_0} \right)^{1/4} G_L^{-1/2} V_p^{-1/4}$$  \hspace{1cm} (12)

Compared with the models of Kurz and Fisher, the coefficient in present simulation is $5.3$.

3 Conclusions

A solute diffusion controlled solidification model was used to simulate the microstructural evolution of Ti-45at.%Al alloy during directional solidification. The following conclusions can be drawn:

(1) At the same pulling velocity, increasing thermal gradient decreases the final primary dendrite spacing, especially at high thermal gradients. Due to branching mechanism in the constrained dendritic growth, the secondary dendrite arms cannot be very well developed in many cases.

(2) A transient structure composed of cells and dendrites was observed during directional solidification. In cell/dendrite transition region, the range of the pulling velocity varies with the thermal gradient, and the higher the thermal gradient, the smaller the range. The simulated results agree reasonably well with experiment results at low pulling velocity.

References