Phase-field simulation of secondary dendrite growth in directional solidification of binary alloys

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Abstract: Phase field method was used to simulate the effect of grains orientation angle θ_{11} and azimuth θ_A of non-preferentially growing dendrites on the secondary dendrites of preferentially growing dendrites. In the simulation process, two single-factor influence experiments were designed for columnar crystal structures. The simulation results showed that, when $\theta_{11} < 45^\circ$ and $\theta_A < 45^\circ$, as θ_{11} was enlarged, the growth direction of the secondary dendrites on the preferentially growing dendrites at the converging grain boundary (GB) presented an increasing inclination to that of preferentially growing dendrites; with increasing θ_A , the growth direction of the secondary dendrites on the preferentially growing dendrites at the converging GB exhibited greater deflection, and the secondary dendrites grew with branches; the secondary dendrites on the preferentially growing dendrites at diverging GBs grew along a direction vertical to the growth direction of the preferentially growing dendrites. When $\theta_A = 45^\circ$ and $\theta_{11} = 45^\circ$, the secondary dendrites grew in a direction vertical to the growth direction of preferentially growing dendrites. The morphologies of the dendrites obtained through simulation can also be found in metallographs of practical solidification experiments. This implies that the effect of a grain's orientation angle and azimuth of non-preferentially growing dendrites on the secondary dendrites of preferentially growing dendrites does exist and frequently appears in the practical solidification process.

Key words: phase-field method; binary alloy; directional solidification; secondary dendrites

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Directional solidification is used to obtain columnar crystals with specific orientations using forced measures in the solidification process ^[1]. Directional solidification enables arrangement of solidified structures of materials along a specific direction to obtain directional and monocrystalline structures, which can improve mechanical and physical properties of the alloy ^[2]. At present, directional solidification has been widely used in aviation and aerospace industries, superconducting, semiconductors, magnetic materials, and in situ composites ^[3-5]. For this reason, directional solidification has become a research focus since the 1970s ^[6-8].

Numerical simulation of microstructures is an effective tool for predicting the morphology of structures formed in directional solidification. Currently, commonly used methods for the numerical simulation of microstructures include cellular automation ^[9-10] and the phase-field

method ^[11-13]. Therein, the phase-field method can couple nucleation model and growth model with its influencing factors on nucleation and growth, along with the basic heat transfer equation, and therefore allows accurate numerical simulation of microstructures ^[14-17]. Scholars have applied the phase-field method to investigate the competitive growth of dendrites in the directional solidification process in recent years. Tomohiro Takaki et al. ^[18] simulated the competitive growth of dendrites and the grain selection of Al-Cu binary alloys in the directional solidification process at converging grain boundaries (GBs) using the 2D phase-field model. Moreover, they summarized the influences of cellular structures and side-branches on dendritic structures upon the fluctuations of divergent GBs. By using the 2D phase-field method and carrying out tests on thin specimens, Chunwen Guo, et al. ^[19] investigated the secondary and tertiary branches at a divergent GB between two columnar crystals with different grain orientations. However, solidification occurs in 3D conditions, so simulation under 2D conditions fails to describe the competitive growth of dendrites in the directional solidification process. To better investigate

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the competitive growth process of dendrites in the directional solidification, some scholars researched the competitive growth of columnar crystals under 3D conditions. For example, Tourret et al.^[7] simulated the competitive growth of monolayer columnar crystals in bicrystalline samples using the 3D phase-field method. They revealed the influences of azimuth on the orientation selection of GB between monolayer columnar crystals under conditions with specific orientation angles of grains. It can be found from the aforementioned studies that side-branches on columnar crystals exert influences on both the selection of GB orientation and the spacing of primary dendrites, and play a significant role in the competitive growth of columnar crystals. However, 3D simulation analysis is seldom applied to the side-branches of columnar crystals. In our previous research^[20], a 3D multi-phase field model of directional solidification was established to simulate the directional solidification process of Al-2% mole-Cu binary alloy, and the influence of grain orientation on competitive growth of dendrites under different competitive modes was studied. However, the effect of grain orientation angle θ_{11} and azimuth θ_A of non-preferentially growing dendrites on the secondary dendrites of preferentially growing dendrites is not considered. The grain orientation angle and azimuth have a great influence on the growth morphology of dendrites during 3D solidification^[7].

In the present research, the same 3D multi-phase field model of directional solidification as in Ref.^[20] was used to study the effect of grains orientation angle and azimuth of non-preferentially growing dendrites on the secondary dendrites of preferentially growing dendrites, and the simulation results were verified by experiments, which can be used as a reference for the actual production.

1 Phase-field method

1.1 Phase-field controlling equation

The phase-field controlling equation^[12, 21] is:

$$\frac{\partial \phi}{\partial t} = M(\varepsilon(\theta)^2 \nabla^2 \phi + (\varepsilon(\theta) \cdot \varepsilon''(\theta) + \varepsilon'(\theta)^2) \cdot (|\nabla \phi| \nabla \bar{n}) - f_\phi) \quad (1)$$

where, ϕ , M and t refer to the order parameter of phase field, migration rate of solid-phase interface, and time variable, and $\varepsilon(\theta)$ refers to a parameter related to interfacial energy. Moreover, f_ϕ represents the first derivative of free energy density on order parameter of phase field.

1.2 Solute-field controlling equation

The solute-field controlling equation^[20] is expressed as follows:

$$\frac{\partial c}{\partial t} = \nabla \cdot \left(\frac{D(\phi)}{f_{cc}} \nabla f_c \right) \quad (2)$$

where, $D(\phi)$, f_c and f_{cc} refer to diffusion coefficient of solutes, the first and second derivatives of free energies on concentration, respectively.

2 Phase-field parameters and numerical calculation of models

Taking Al-2% mole-Cu alloy as an example, the competitive growth of dendrites in directional solidification process was investigated. The physical parameters of the alloy are listed as in Ref.^[20].

Equations (1) and (2) are calculated simultaneously by using the explicit finite difference method, both of which apply the same time step Δt , where Δt and special step Δx ($\Delta x = \Delta y = \Delta z$) satisfy the following stable conditions:

$$\Delta t < \min \left\{ \Delta x^2 / (6D_L), \Delta y^2 / (6D_L), \Delta z^2 / (6D_L) \right\} \quad (3)$$

where, D_L refers to the diffusion coefficient of liquid-phase solute.

The simulation range, initial placement of grains (as shown in Fig. 1), establishment of the sub-cooled melt temperature field, and the setting of other boundaries are the same as in Ref.^[20].

Fig. 1: Placement of initial nuclei

Figure 2 shows the orientation angle and the azimuth of columnar crystals. Where θ_A is the azimuth angle and θ_{11} (denoting the projection of the included angle between the grain orientation and the z-axis on the xoz-plane) is the grain orientation angle. Suppose that the dendrites whose growth direction was parallel to the temperature gradient (green arrows in Fig. 2) were preferentially growing dendrites, and those with an angle of θ_{11} with the temperature gradient were non-preferentially growing dendrites. And suppose that grains 1, 4, 5, and 7 in Fig. 1 were preferentially growing grains with both θ_{11} and θ_A of 0° , while grains 2, 3, and 6 were non-preferentially growing grains with azimuths of θ_A . Between preferentially growing dendrite 5, and non-preferentially growing dendrite 2, a diverging GB was formed, while converging GBs were found between preferentially growing dendrite 7 and non-preferentially

Fig. 2: Orientations of dendrites

growing dendrites 2, 3, and 6, as illustrated in Fig. 2. The grains grew along the direction indicated by the black arrows in Fig. 2.

3 Results and discussion

3.1 Simulation results and analysis of columnar crystals

To describe the influences of θ_A and θ_{11} on the secondary

dendrites on the preferentially growing dendrites, the following two single-factor influence simulations were conducted: (1) θ_A remained unchanged to investigate the influences of θ_{11} on the secondary dendrites on the preferentially growing grains. (2) θ_{11} was fixed to study the influences of θ_A on the secondary dendrites on the preferentially growing grains.

Figure 3 shows the simulated morphologies in competitive growth of two-layer columnar crystals at 10,000 Δt with

Fig. 3: Morphologies of competitive growth of dendrites with different combinations of θ_A and θ_{11}

combinations of different θ_A and θ_{11} . It can be seen that as the orientation angles of non-preferentially growing dendrites varied, the degree of interaction between dendrites also changed, as indicated by the different degrees of inhibition on non-preferentially growing dendrites 2, 3, and 6. The non-preferentially growing dendrites in Figs. 3 (a1-e1), (a2-e2), (a3-e3), and (a4-e4) were inhibited and stopped growing in succession.

Figure 3 shows the 3D external morphologies of the columnar crystals from the certain perspective. It can be seen that the 3D morphologies are mutually blocked, thus influencing the

observation of the change in the secondary dendrites on the preferentially growing dendrites. Therefore, to reveal the results of the two single-factor simulations, Fig. 3 was sliced. In this way, the changes in the secondary dendrites and their effect on the preferentially growing dendrites with changes in θ_{11} of the non-preferentially growing dendrites can be directly observed from the slices parallel to the xoz -plane, as shown in Fig. 4. Similarly, to observe the variation of the secondary dendrites on the preferentially growing dendrites with changes in θ_A of the non-preferentially growing dendrites, Fig. 3 was sliced parallel to the xoy -plane, as shown in Fig. 5. It shows the top views,

Fig. 4: Morphologies of competitive growth of dendrites with different combinations of θ_A and θ_{11} (slices parallel to xoz -plane)

Fig. 5: Top views and morphologies of competitive growth of dendrites under combinations of different θ_A and θ_{11} (slices parallel to the xoy -plane)

the phase field slices figure and the solute field slices figure corresponding to each 3D result.

Secondary dendrites grew on the preferentially growing dendrite 7 when the θ_A and θ_{11} values of the non-preferentially growing dendrites were 0° and 7.5° . Meanwhile, they grew

along the direction vertical to the growth direction of the non-preferentially growing dendrites. The non-preferentially growing dendrite 3 was inhibited by the preferential dendrite 7 and stopped growing, as shown in Fig. 4 (b). As θ_{11} increased, it can be observed that the growth direction of secondary dendrites

on preferentially growing dendrite 7 inclined more than that of the preferentially growing dendrite [(white arrows in Figs. 4 (b1–b4)]. The secondary dendrites on preferentially growing dendrite 5 grew along the direction vertical to the growth direction of dendrite 5, as indicated by yellow arrows in Figs. 4 (b1–b4). At the same time, the secondary dendrites on the preferred dendrites 5 and 7 have an enhanced inhibitory effect on the non-preferred dendrites 2, 3 and 6, which causes the non-preferred dendrites 2, 3 and 6 to stop growing in succession, as shown in Fig. 4 (b1–b4). When θ_A was 10°, 20°, 30°, and 45°, the secondary dendrites on preferentially growing dendrites 5 and 7 showed the same trend in growth as that when θ_A was 0° with increasing θ_{11} , as shown in Figs. 4 (c–c4), (d–d4), (e–e4), and (f–f3). This is because when θ_A was unchanged, the preferentially growing dendrites inhibited the growth of their non-preferentially growing counterparts with changes in θ_{11} . At the same time, they occupied the liquid-phase space in the growth front of non-preferentially growing dendrites and therefore produced secondary dendrites. Meanwhile, the solute atoms released by the non-preferred dendrites at the growth front were enriched, and the larger the value of θ_{11} is, the greater the degree of enrichment, such that the growth direction of the secondary dendrites is more inclined. Moreover, the secondary dendrites on preferentially growing dendrite 7 in Fig. 4 (f4) grew vertically to the growth direction of the preferentially growing dendrites, as indicated by the white arrows in Fig. 4. This is determined by the relative positions of grains in the two-layer columnar crystals: when $\theta_A = 45^\circ$ and $\theta_{11} = 45^\circ$, secondary dendrites on preferentially growing dendrites at converging GBs rapidly occupied the liquid-phase space in the growth front of non-preferentially growing dendrites, so their growth was weakly inhibited by solute atoms in the growth front of the non-preferentially growing dendrites.

If the θ_{11} and θ_A values of non-preferentially growing dendrites were 7.5° and 0°, the growth direction of secondary dendrites on the preferentially growing dendrite 5 was deflected, as indicated by the white arrow in Fig. 5 (b). When θ_{11} remained unchanged, the growth direction of the secondary dendrites on preferentially growing dendrite 7 was deflected more with the increase of θ_A as indicated by the black arrows in Figs. 5 (b–f). The occurrence of bifurcation growth of secondary dendrites is evident, as indicated by the black arrows in Figs. 5 (e–f).

In addition, the secondary dendrites on preferentially growing dendrite 5 were deflected in the growth direction, as indicated by the white arrows in Figs. 5(b–e). As θ_A was increased to 45°, the growth direction of the secondary dendrites on preferentially growing dendrite 5 was vertical to that of the preferentially growing dendrite, as demonstrated in Fig. 5(f).

When θ_{11} was 10°, 15°, 30°, and 45°, the secondary dendrites on preferentially growing dendrite 7 showed the same trend in their growth as that when the θ_{11} was 7.5° with increasing θ_A , as indicated by black arrows in Figs. 5 (a1–f1), (a2–f2), (a3–f3), and (a4–e4); meanwhile, the growth direction of secondary dendrites on preferentially growing dendrite 5 was vertical to that of the preferentially growing dendrites (Figs. 5 (a1–f1), (a2–f2), (a3–f3) and (a4–e4)). This is because when θ_{11} remained unchanged, non-preferentially growing dendrite 3 exerted weak inhibition effects on preferentially growing dendrite 7 and the direction of growth of the secondary dendrites on preferentially growing dendrite 7 was deflected slightly when θ_A was 0°. As θ_A increased, it took less time for non-preferentially growing dendrite 3 to reach the growth front of preferentially growing dendrite 7 (Fig. 6). The released solute atoms accumulated at the growth front of the secondary dendrites on preferentially growing dendrite 7 (as indicated by the white circles in Fig. 6), thus inhibiting the normal growth of secondary dendrites on preferentially growing dendrite 7 and deflecting their direction of growth. Figure 6 shows the phase field and solute field distribution of grains positions in columnar crystal with different θ_A , and the color corresponding to the value of the solute field order parameter is shown as the bar. If θ_A further increased, the degree of inhibition increased, so that secondary dendrites on preferentially growing dendrite 7 formed branches. In contrast, as preferentially growing dendrite 5 was slightly inhibited, the secondary dendrites thereon basically retained the same growth direction. The reasons for the deflection of the growth direction of secondary dendrites on preferentially growing dendrite 5 in Figs. 5 (b–e) is that the solute atoms released by non-preferentially growing dendrites in the periphery during growth influenced the preferentially growing dendrites at diverging GBs, as indicated by red circles in Figs. 5(b–e). The reasons for the growth of secondary dendrites on preferentially growing dendrites vertical to that of the preferentially growing dendrite in Fig. 5 (f4) are the same as above.

Fig. 6: Positions of grains in the columnar crystals with different θ_A

3.2 Experimental result

The Al-2% mole-Cu was prepared by directional solidification experiment to verify the phase field simulation results. Sample preparation was carried out in the same manner as in Ref. [20]. The experimental results are shown in Fig. 7. It can be found that the morphology is similar to the growth morphology

appearing in the slice of the simulation result. For example, the morphology marked using the red boxes in Figs. 7 (a–c) was similar to that in Figs. 5 (b2–f4). Additionally, the morphologies marked using yellow circles in Figs. 7 (e–f) was similar to that in Figs. 4 (b1–b4).

Fig.7: Directional solidification microscopic morphology of Al-2% mole-Cu

4 Conclusions

(1) When $\theta_{11} < 45^\circ$ and $\theta_A < 45^\circ$, as θ_{11} increases, the growth direction of secondary dendrites on preferentially growing dendrites at converging GBs inclines more significantly to that of the preferentially growing dendrites; while the secondary dendrites on preferentially growing dendrites at diverging GBs grow along the direction vertical to that of the preferentially growing dendrites. With increasing θ_A , the growth direction of secondary dendrites on preferentially growing dendrites at converging GBs is deflected more significantly and the bifurcation growth of secondary dendrites occurs at diverging

GBs; the growth of secondary dendrites on preferentially growing dendrites is vertical to that of the preferentially growing dendrites.

(2) When $\theta_A = 45^\circ$ and $\theta_{11} = 45^\circ$, the secondary dendrites on preferentially growing dendrites at converging GBs present different growth trends. The secondary dendrites grow in a direction vertical to the growth direction of preferentially growing dendrites.

(3) Experimental results are consistent with the simulated results, proving that the effect of grains orientation angle and azimuth of non-preferentially growing dendrites on the secondary dendrites of preferentially growing dendrites do exist and frequently appear.

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