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# Assessment of scaling factor in modified dendrite growth model ①

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**[Abstract]** A model for dendrite growth during rapid solidification was established on the basis of BCT model and marginal stability criterion through modified Peclet numbers. Taking into account the interaction of diffusion fields, including solute diffusion field and thermal diffusion field around the dendrite tip, the model obtain a satisfactory results to predict the dendrite velocity and the tip radius, which agrees well with the experimental data from references in Cu-Ni alloy.

**[Key words]** rapid solidification; undercooled melt; dendrite growth; Cu-Ni alloy
**[CLC number]** TG 146; TG 114 **[Document code]** A

## 1 INTRODUCTION

Rapid solidification (RS) processing is now well established for producing materials with metastable and refined microstructures and improved properties. RS is a solidification process with high growth velocity (1~ 100 cm/s) obtained by the rapid rate of melt cooling, high initial nucleation undercooling or by the rapid moving of temperature field[1]. Constitutional effects of RS include extension of solid solubility and the formation of metastable phases due to nucleation and growth competition between the candidate phases in the undercooled melt.

For rapid solidification, many theories[2~6] about nucleation and growth were put forward to analyze the solidification behavior and the microstructure formation. In the present paper, we'll pay attention to the dendrite growth phenomenon.

For the dendrite growth in undercooled melt, many theoretical models[3~6] based on the Ivantsov solutions for the thermal and solute diffusion fields, negative temperature gradient ahead of the tip and the marginal morphological stability criterion of the solid/liquid interface, have been established to understand isolated dendrite growth behavior. Accordingly, experiments of dendrite growth in rapid solidification[7~10] have confirmly supported the current dendrite growth theories.

For the directional solidification, a model referred to the Laxmanan's model[11,12], was developed with a positive thermal gradient ahead of the dendrite tip. While in undercooled melt, the phenomenon of directional growth of dendrites have also been observed in practice[13]. Thus, on the assumption that the diffusion fields around the dendrite tips being overlapped, a directional dendrite growth model in undercooled melt has been developed by LI et al[13] on the basis of BCT model and marginal stability criterion.

The LI's model incorporated different forms to express the balance equations. As for temperature distribution, it is $P_t = \Omega_t$ which is equivalent to the Ivantsov solution; while for solute distribution, it is $\Omega_c = Iv(P_c)$ which is equivalent to the spherical solution. During deduction of the mass and heat transfer balance equations, there was also a mistake probably occurred in the balance equations. As for mass transfer, the balance equation $vc_l^* (1- k) = D_l G_c$ ought to be multiplied by two on the right side, and then be replaced by

$$vc_l^* (1- k) = - 2D_l G_c \quad (1)$$

Similarly, the heat transfer balance equation $v \Delta H = K_l G_l$ should also be corrected by

$$v \Delta H = 2K_l G_l \quad (2)$$

In order to solve such problems mentioned above, we have developed a modified model for dendrite growth in undercooled melt based on BCT model[6] and marginal stability criterion[14~17] with unified assumption of a dendrite with parabolic revolution shape, in which both mass and heat distributions ahead the dendrite tips accord well with Ivantsov solution through modified Peclet numbers.

## 2 MODIFIED DENDRITE GROWTH MODEL

During the growth of dendrites, especially for the constrained dendrites in an undercooled melt, the released solute and latent heat of fusion may interact through the overlapping of the diffusion fields[13]. Such a dendrite growth mode is so sophisticated that the exact mathematical solution for this mode of dendrite growth is very difficult. Therefore, we adopt an analytical way similar to Laxmanan's treatment[11,12]. The method combines the interaction of dendrites into a factor λ to describe the diffusion

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fields related to the tip radius as $\delta = \lambda R$, where $\lambda$ is a scaling factor defined as a ratio between $\delta$ and $R$ to express the interaction of the dendrites, $\delta$ is the effective diffusion thickness, $R$ is the tip radius.

Firstly, we give some assumptions that during the process of solute transfer in a binary alloy, any chemical reaction is excluded, and neglect the diffusion in solid, the heat convection in the liquid and the temperature dependence on the material parameters. Thus, through the solute balance Eqn. (1), we can obtain

$$vc_l^*(1-k) = -2D_l\left(\frac{dc}{dr}\right)_{\delta_c} \quad (3)$$

where $\left(\frac{dc}{dr}\right)_{\delta_c}$ is the solute gradient ahead of the tip, which can be described as

$$\left(\frac{dc}{dr}\right)_{\delta_c} = -(c_l^* - c_0)/\delta_c \quad (4)$$

Substituting Eqn. (4) into Eqn. (3), we can obtain

$$\frac{v\cdot\delta_c}{2D_l} = \frac{c_l^* - c_0}{c_l^*(1-k)} \quad (5)$$

From the assumption $\delta_c = \lambda_c \cdot R$, Eqn. (5) can be revised as

$$\frac{v\cdot\lambda_c\cdot R}{2D_l} = \frac{c_l^* - c_0}{c_l^*(1-k)} \quad (6)$$

or

$$\lambda_c\cdot P_c = \Omega_c \quad (7)$$

where $P_c = vR/2D_l$ is the solute Peclet number, $c_l^*$ is the solute concentration at the dendrite tip.

As for dendrite growth controlled by thermal diffusion, we can obtain a conclusion on the assumption of $\delta_t = \lambda_t \cdot R$ as

$$\frac{v\cdot\lambda_t\cdot R}{2\alpha_l} = \frac{T_l^* - T_\infty}{-\Delta H_m/c_p} = \Omega_t \quad (8)$$

or

$$\lambda_t\cdot P_t = \Omega_t \quad (9)$$

where $P_t = vR/2\alpha_l$ is the thermal Peclet number, $\Delta T_t = T_l^* - T_\infty$ is the thermal undercooling.

Note, in calculating the concentration gradient at the tip in dendrite growth models proposed earlier, it was assumed that the diffusion distance in front of the tip is equal to the dendrite tip radius $R$, and then the solute gradient always be regarded as $\left(\frac{dc}{dr}\right)_{\delta_c} = \left(\frac{dc}{dr}\right)_R = -(c_l^* - c_0)/R$ (or $\left(\frac{dT}{dr}\right)_{\delta_t} = \left(\frac{dT}{dr}\right)_R = -(T_l^* - T_0)/R$ for thermal gradient) in simplicity. As we have discussed above, the diffusion distance might be different from $R$ due to the interaction of dendrites and other factors and should be replaced by $\lambda R$, where $\lambda$ is a positive quantity, which will be discussed in section 4.

On the basis of the assumption above, for dendrite growth controlled by solute diffusion, the modified thermal peclet number can be described as

$$P'_c = \lambda_c\frac{vR}{2D_l} \quad (10)$$

where $D_l$ is the solute diffusivity in liquid. It can be explained by the ratio of the dendrite radius $R$ to the thickness of diffusion fields $\delta$. For this reason, the value of dendrite growth velocity $v$ must be decreased due to interaction of diffusions, however, compared with isolated dendrite, the total value of $\delta$ should be increased.

According to the assumption of dendrite with parabolic revolution shape, the solute distribution in liquid can be described with Ivantsov solution[4]:

$$\Omega_c = Iv(P'_c) \quad (11)$$

where $\Omega_c$ is defined as the dimensionless solute supersaturation.

On the similar assumption, for dendrite growth controlled by thermal diffusion, the modified thermal peclet number can be described as

$$P'_t = \lambda_t\frac{vR}{2\alpha_l} \quad (12)$$

where $\alpha_l$ is the thermal diffusivity in liquid.

And the thermal distribution around the tip in liquid can be described with Ivantsov solution:

$$\Omega_t = Iv(P'_t) \quad (13)$$

where $\Omega_t$ is defined as the dimensionless undercooling.

According to the dendrite growth model of BCT[6], the dendrite tip undercooling can arise from four sources, i. e. requirements for diffusion of heat and of solute from growing tip, the effect of interface curvature and interface kinetics. Thus the total undercooling $\Delta T$ consists of four contributions: the thermal undercooling $\Delta T_t$, the solute undercooling $\Delta T_c$, the curvature undercooling $\Delta T_r$ and the kinetic undercooling $\Delta T_k$.

$$\Delta T = \Delta T_t + \Delta T_c + \Delta T_r + \Delta T_k \quad (14)$$

The forms of the four undercoolings are similar to that from BCT model except that the Peclet number should be modified.

Accordingly, the marginal stability criterion[14~17] can also be expressed through modified Peclet numbers as

$$R = \frac{\Gamma/\sigma^*}{\frac{P'_t L}{c_p}\zeta_t - \frac{2m'P'_c c_0(1-k)}{1-(1-k)Iv(P'_c)}\zeta_c} \quad (15)$$

where

$$\sigma^* = \frac{1}{4\pi^2} \qquad \zeta_t = 1 - \frac{1}{\sqrt{1+\frac{1}{\sigma^* P'^2_t}}}$$

$$\zeta_c = 1 + \frac{2k}{1-2k-\sqrt{1+\frac{1}{\sigma^* P'^2_t}}}$$

## 3 MODEL RELIABILITY

In order to prove the reliability of this modified model, we selected some experimental data measured by Herlach and LI et al[10, 13] to compare with the results calculated by this model. From LI's discussion, we know that $\lambda_c \ll \lambda_t$ and we'll only discuss the role of $\lambda_t$. Using the physical parameters of $Cu_{70}Ni_{30}$ listed in Table 1, and assigning different values to $\lambda_t$, we've obtained the relationship of growth velocity and the corresponding undercooling in Fig. 1. By comparison, the measured data of dendrite growth velocity and undecooling were also marked. In Fig. 1, the curves 4, 5, 6, 7 are calculated with constant values respectively as $\lambda_t = 1$, $\lambda_t = 1.5$, $\lambda_t = 2.0$, and $\lambda_t = 2.5$, and the curve 3 is calculated with relationship $\lambda_t = 2.5 - 0.4v$, the curve 2 is calculated by Herlach[10] and curve 1 by LI[13]. From Fig. 1, it can be seen that the curve calculated with relationship agrees better with the experimental data than the results calculated by Herlach and LI.

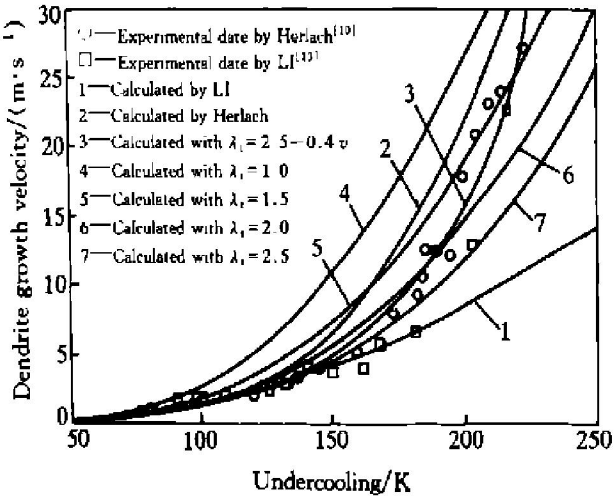


**Fig. 1** Relationship between dendrite growth velocity and undercooling calculated by different models with experimental data

**Table 1** Physical properties of $Cu_{70}Ni_{30}$ alloy[10~13]

| Parameter | $\Delta H/(J\cdot mol^{-1})$ | $c_p/(J\cdot mol^{-1}\cdot K^{-1})$ | $D_l/(m^2\cdot s^{-1})$ |
|---|---|---|---|
| Value | 13 940 | 31.436 | 4.73 |
| Parameter | $\alpha_l/(m^2\cdot s^{-1})$ | $\Gamma/(K\cdot m)$ | $M/(kg\cdot mol^{-1})$ |
| Value | $40\times 10^{-6}$ | $2.627\times 10^{-7}$ | 0.062 |
| Parameter | $\rho/(kg\cdot m^{-3})$ | $k_0$ | $m_l/(K\cdot \%^{-1})$ |
| Value | 8 900 | 1.485 | 4.567 |
| Parameter | $v_0/(m\cdot s^{-1})$ | $a_0/m$ | $T_l/K$ |
| Value | 2 000 | $3.0\times 10^{-10}$ | 1 513.48 |

## 4 DISCUSSION

### 4.1 Solute and thermal diffusion field

The thermal diffusivity is generally about three orders of magnitude greater than the solute diffusivity, so that the thermal diffusion length $l_t$ in the liquid must be much longer than the solute diffusion length $l_c$[13]. Especially for rapid solidification, the marked solute trapping[18~21] confined the solute diffusion to a narrow region around the tip. Accordingly to Laxmanan[11, 12], $\lambda_t$ is much larger than $\lambda_c$ due to the great difference between the thermal diffusivity and solute diffusivity in liquid, especially for rapid solidification.

To get an insight, a comparison between the solute diffusion length $l_c$, and the thermal diffusion length $l_t$ is made as follows. The solute diffusion length in the liquid in the growth direction is

$$l_c = \delta_c = \frac{D_l}{v} \tag{16}$$

The thermal diffusion length in the liquid in the growth direction is

$$l_t = \delta_t = \frac{\alpha_l}{v} \tag{17}$$

Now, let $v = 1\sim 10\,m/s$, $R$ is in the range of $(1\sim 5)\times 10^{-7}\,m$, $l_c$ is about the same order of $D_l$, $10^{-10}\,m$ or so, $l_t$ is about the same order of $\alpha_l$, $10^{-6}\,m$ or so.

According to the orders of diffusion length and the value $R$, it can be deduced that the solute diffusion around the tip is confined to a very narrow region, and that the solute diffusion interaction among the dendrites can be ignored[13]. As far as we know from LI's discussion, the stronger the interaction between the dendrites is, the thicker the diffusion field in the liquid consequently becomes. Thus with the increasing of $\lambda$, the diffusion length becomes longer, the dissipation of latent heat (or solute) becomes more difficult and the dendrite growth velocity decreases.

In summary, from the analysis above, we can obtain that the value of $\lambda$ being equaled to unity means that the effect of interaction through the overlapping of the diffusion fields could be ignored, which accords to the growth mode of isolated dendrite. The value $\lambda_c = 1$ and $\lambda_t > 1$ means that the interaction of solute diffusion can be neglected, which usually applied for growth of constrained dendrites only controlled by thermal diffusion. While the values $\lambda_c > 1$, $\lambda_t = 1$ mean that the interaction of thermal diffusion can be neglected, which usually applied for growth of constrained dendrites only controlled by solute diffusion.

### 4.2 Comparison of curves with different $\lambda_t$

Fig. 2 shows a series of curves between $v$ and $R$ calculated with different $\lambda_t$ value in the case of Cu-Ni alloy from Eqns. (14) and (15). From Fig. 2, we can know that at the same velocity, the dendrite tip radius calculated with higher $\lambda_t$ is bigger than the case of lower $\lambda_t$. Meanwhile from Fig. 1, we can also know that for rapid solidification, the required undercooling with higher $\lambda_t$ is smaller than that with lower $\lambda_t$ at equal velocity. From above, it can be concluded that with increasing $\lambda_t$, the influence of thermal interaction between the dendrites on the growth velocity becomes more remarkable, which in principle, agrees with our deduction. In addition, the curves calculated with $\lambda_t = 2.5 - 0.04V$ are more flat and smooth than the curves with constant $\lambda_t$, but the characteristics are similar.

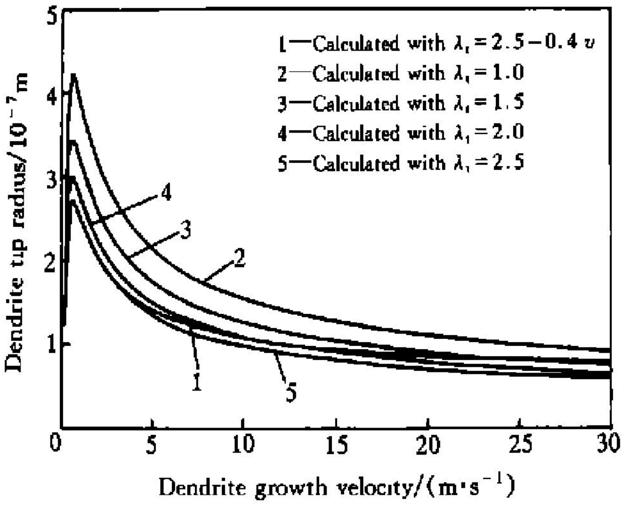


**Fig. 2** Curves of dendrite radius and dendrite growth velocity with different $\lambda_t$( $\lambda_c$= 1)

### 4.3 Relationship between $\lambda$ and $V$

In the present model, we firstly consider the scaling factor $\lambda_t$ as constant, and the calculated results are depicted by curves 4, 5, 6, 7 in Fig. 1. Compared with the experimental data, it can be seen that under lower undercooling (about 50~ 140K), the experimental data fit the model better with higher value of $\lambda_t$, while under higher undercooling (larger than 180K), the more appropriate value of $\lambda_t$ is lower. Accordingly, we think that there must be some factors that influence the scaling factor $\lambda_t$. From Kurz[17] and Aziz[18~ 21], we know that in rapid solidification, with increasing growth velocity, the solute trapping and the thermal dragging increase remarkably, thus these effects will decrease the solute enrichment and the heat accumulation at the dendrite tip, that is to say it will shorten the diffusion length of the solute and the heat. In addition, during the mathematical deduction, many assumptions such as neglecting of the heat convection in the liquid and the temperature dependence on the material parameters have been made in order to simply the calculation, but all these factors are evident in the case of RS. Furthermore, there are also many other factors, which can not be directly described. Therefore, we should find a relationship to consider all these factors which are difficult to express analytically. In the present modified model we approximately define the scaling ratio $\lambda$ as a function of dendrite growth rate by a linear relationship $\lambda = -\alpha V + \lambda_0$, which is called as modified factor determined by the effects of all the factors mentioned above, $\lambda_0$ is an extrapolating value when the growth velocity approaches to zero.

Besides, it must be emphasized that for rapid solidification, neglecting the solute diffusion interaction means that the dendrite growth velocity is mainly controlled by thermal diffusion. However, at low growth velocity in conventional solidification and rapid solidification with the growth velocity below 100 cm/s(not deeply undercooled), the solute interaction can not be neglected, therefore another relationship about ought to be given.

## 5 CONCLUSIONS

1) A modified model for the dendrite growth in undercooled melts has been established using the Ivantsov solution both for solute distribution and for the temperature distribution ahead the tip, and also incorporating the interaction between the dendrites through the scaling factor $\lambda$.

2) When the dendrite growth velocity increases with the increasing undercooling, the scaling factor $\lambda$ should decrease for some reasons. An additional factor has been employed to express such effects on scaling factor $\lambda$ through a linear relationship with the velocity as $\lambda = \lambda_0 - \alpha v$.

3) The experimentally measured dendrite growth velocity in undercooled Cu-Ni alloy is in a good agreement with the results calculated by the present model with the relationship of $\lambda_t = -0.4v + 2.5$. It is apparent that with suitable value of $\lambda_0$ and $\alpha$, consistent results can be obtained for any undercooled alloy systems.

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