Simulation for microstructure evolution of Al-Si alloys in solidification process

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Abstract. In this paper, two phase field models for free interface of dendrite growth as sharp interface model and diffusion interface have been introduced; and microstructure evolution in solidification process is studied by phase field model, model of the PFM and its numerical solution are introduced, the dendrite growth process of Al-Si alloy in solidification process is simulated, and the simulation results has been analyzed, The comparisons of dendrite morphology between simulated results and investigations by others are also presented, which proved that the dendrite morphology are similar in trunks and arms growth, so proved the developed phase field program is accurate. But this study is still at the stage of theoretical exploration, and it is necessary to be linked with engineering application further.

1. Introduction
   Early in 1966, Oldfield et al. [1] proposed that the heat source term in heat transfer equation could be represented by the function of nucleation rate and growth velocity of crystal grain, and he attempted to simulate the solidification microstructure of gray casting iron. Yet the micro simulation had been developed slowly confined by the corresponding macro simulation during followed decades. Currently, there mainly exist three kinds of models: Deterministic Model, Stochastic Model and Phase-field Model. In this paper, author adopted phase-field model, and in this model the phase field variable \( \varphi \) was introduced to describe the morphology, curvature and movement of phase interface. Coupled with thermal field, solute field, flow field and other outside fields, phase field model can simulate the real solidification process.
   Since 1980s, Collins [2], Langer [3] and Caginalp[4] proposed the initial phase field model. Then, Caginalp [5-7], Fife and Gill [8, 9] suggested that when the interface width is near zero, PFM would be the sharp interface model, and they firstly introduced the anisotropic factor. Fix [10] carried out the numerical solution of phase field equations for the first time. In 1993, Kobayashi [11] realized the 2-D simulation for the dendrite growth in undercooling pure melts by this model coupled with anisotropic influence. Wheeler and Murry [12] quantitatively simulated the solidification microstructure of pure Ni through the thermodynamically consistence phase field model proposed by Wang [13] and Penrose [14]. In 1993, Wheeler [15] developed the binary alloy phase field model, and predicted the solute micro segregation and the dendrite morphology. In1996, Karma and Rappel[16,17]obtained effective Gibbs-Thomson dependence within certain interface width, and suggested that the interface width could thicker than capillary length, so developed a new phase field model, which can simulate deep
undercooling condition and shorten the calculation time. Provatas [18] successfully simulated the single crystal grain morphology through the self-fitted grid technology. Steinbach [19] first proposed the phase field concept of multi phase system and deduced the governing equations. Nestler [20] present adaptive finite element simulations of dendritic and eutectic solidification in binary and ternary alloys. Two-dimensional phase-field simulations are used by J.C. Remirez and C. Beckermann [21] to test standard theories for free dendritic growth of alloys.

Phase field method model become the research hotspot within modeling material science as soon as it has been proposed, so it is of great potential and importance [22-25].

2. Numerical model of PFM (Phase Field Model)

At present, there are such two models for free interface of dendrite growth as sharp interface model and diffusion interface Model.

Sharp interface model is shown in Fig 1, for an close space $\Omega$, the simulated area was divided into solid states $\Omega_+(t)$ and liquid state $\Omega_-(t)$ by the sharp interface $\Gamma(t)$, across which the system changes suddenly from solid state to liquid state and $\Gamma(t)$ is taken as no thickness. So, temperature and material properties parameters of the two sides across the interface are un-continuous.

In diffusion interface model (shown in Fig 2), solid/liquid interface has a certain thickness, and various transmission phenomena are completed at this interface area. Solid phase, liquid phase and the interface are taken as different phases represented by a scalar, which can form a group of differential equations together with other variables. Therefore, it need not to track interface and separate liquid and solid phase, whereas, use a scalar to track the phase in system. Thus, the same numerical method can be adopted in the entire simulated zone, and can raise the calculation precision. Moreover, compared to the sharp interface model, the diffusion interface is more close to the physical fact, so it has been widely applied in various phase transformation process.

Diffusion interface model is adopted in PFM, the phase field variable $\Phi$ is introduced, which addresses the physical state (liquid or solid state) of the system with respect to time and space. $\Phi = 1$ represents solid phase, $\Phi = 0$ represents liquid phase, and $\Phi$ changes continuously from 1 to 0 at solid/liquid interface. From Fig 2, we can easily see that there has a diffusion interface layer between...
solid and liquid phase, $\Phi$ changes gradually from a certain constant at one side of the interface to another constant at the other side.

There is

$$\Phi(x,t) = 1 \quad x \in \Omega_+(t)$$

(1)

$$\Phi(x,t) = 0 \quad x \in \Omega_-(t)$$

(2)

Based on Ginzburg-Landau free energy theory [26], Phase field equation could be deduced from the free energy $F$ or entropy $S$, which has been called free energy function method or entropy function method. So the expression of phase field model is different with various address of free energy. In this paper, the fundamental formula of phase field model from free energy $F$ and entropy $S$ of the system will be presented respectively.

According to Ginzburg-Landau free energy theory [26], for a enclosed system with volume $\Omega$, $F$ is:

$$F = \int_{\Omega} \left[ f(\Phi, c, e) + \frac{1}{2} \epsilon^2 (\nabla \Phi)^2 + \frac{1}{2} \delta^2 (\nabla c)^2 \right] d\Omega$$

(3)

Based on the minimum energy principle, the variation of Lyapounov function and the linear un-reversible dynamics, such equations can be deduced as following

$$\frac{\partial \Phi}{\partial t} = -M_{\phi} \frac{\delta F}{\delta \Phi}$$

(4)

$$\frac{\partial c}{\partial t} = \nabla \cdot \left( M_c \nabla \frac{\delta F}{\delta c} \right)$$

(5)

$$\frac{\partial e}{\partial t} = \nabla \cdot \left( M_e \nabla \frac{\delta F}{\delta e} \right)$$

(6)

According to Ginzburg-Landau entropy theory, for the same system as above, the expression of
entropy $S$ is:

$$S = \int_{\Omega} \left[ s(\Phi, c, e) - \frac{1}{2} \varepsilon^2 (\nabla \Phi)^2 + \frac{1}{2} \delta^2 (\nabla c)^2 \right] d\Omega$$  \hspace{1cm} (7)

Such can also be given

$$\frac{\partial \Phi}{\partial t} = M'_{\Phi} \frac{\partial S}{\partial \Phi}$$  \hspace{1cm} (8)

$$\frac{\partial c}{\partial t} = -\left( \nabla \cdot M'_{c} \nabla \frac{\partial S}{\partial c} \right)$$  \hspace{1cm} (9)

$$\frac{\partial e}{\partial t} = -\left( \nabla \cdot M'_{e} \nabla \frac{\partial S}{\partial e} \right)$$  \hspace{1cm} (10)

Where $f$ and $s$ are respectively the free energy density function and entropy density function, $c$ is the alloy composition, $e$ is the internal energy, $\varepsilon$ and $\delta$ are the gradient coefficients of phase field and solute field gradient respectively, $M_{\Phi}, M'_{\Phi}$ are the phase field parameters related to the interface dynamics; $M_{c}, M'_{c}$ are the phase field parameters related to the solute diffusion, $M_{e}, M'_{e}$ are the phase field parameters related to the heat transfer.

For a binary alloy, the model is constructed based on such conditions: (1) Energy and entropy balance equation; (2) The relationship between the thermal-dynamics driving force and field flux of all kinds of exterior field such as temperature field and concentration field follows linear laws; (3) Local entropy is positive.

Formulation (11) is the phase field governing equation proposed by Karma [27].

$$\tau(\theta) \frac{d\Phi}{dt} = \nabla \left[ W^2(\theta)(\nabla \Phi) - g'(\Phi) - Qh'(\Phi)(1-c)H_A + cH_B \right]$$  \hspace{1cm} (11)

Here, $t$ is time, by defining $W(\theta) = W_0 a_s(\theta)$ and $\tau(\theta) = \tau_0 a_s^2(\theta)$, the interface anisotropy is introduced, $W_0$ is the interface thickness, $\tau_0$ represents the time of atomic movement between the solid-liquid interfaces, $a_s(\theta)$ is the factor of anisotropy and $a_s(\theta) = 1 + \gamma \cos(k(\theta_0, \theta))$ , $\theta = \frac{\nabla \Phi}{|\nabla \Phi|}$ denotes the crossing angle between dendrite principal axis and normal direction of interface, $\theta_0=45^\circ$ is its initial value, $\gamma$ and $k$ are the exponent and the modulus of anisotropy respectively. $h(\Phi)$ is a function constructed for the latent heat released during the process of dendrite growth and $g(\Phi)$ is a function constructed for the free energy of the system, $h'(\Phi)$ and $g'(\Phi)$ are the differential coefficients of $h(\Phi)$ and $g(\Phi)$ respectively, $c$ is the concentration of the solute, $Q$ is a balance coefficient.

In equation (11),

$$h(\Phi) = \Phi$$  \hspace{1cm} (12)

$$g(\Phi) = \Phi^2 (1-\Phi)^2$$  \hspace{1cm} (13)

$$H_A = L^A \left( \frac{1}{T} - \frac{1}{T_{mA}} \right)$$  \hspace{1cm} (14)

$$H_B = L^B \left( \frac{1}{T} - \frac{1}{T_{mB}} \right)$$  \hspace{1cm} (15)
Here, $L_A$ and $L_B$ are the latent heat of component A and B, with the melting point $T_{m_A}$ and $T_{m_B}$ respectively.

\[
\frac{dc}{dt} = -\nabla \cdot \left[ M_c h'(\Phi)(H_A - H_B) \nabla \Phi \right] + \nabla \cdot \left( \frac{R}{v_m c(1-c)} M_c \nabla c \right) - \nabla \cdot \left[ M_c \Gamma(\Phi,T) \nabla T \right] - \nabla \left[ M_c \left( \delta^2 \nabla^2 c \right) \right]
\]  

Eq. (16) is the governing equation of concentration, it is assumed that solute diffuse hardly in solid phase, solute distribution is homogeneous in liquid everywhere and solute redistribution only takes place between the solid-liquid interfaces.

Here,

\[
\Gamma(\Phi,T) = \nabla \cdot \frac{2h(\Phi)}{T^2} (L_A - L_B) \]

\[
M_c = \frac{v_m}{R} (1-c)[D_s - h(\Phi)D_L]
\]  

Where $v_m$ is the molar volume, $R$ is the gas constant, $D_S$ and $D_L$ is the solute diffusion coefficient of solid and liquid phase respectively, $\delta$ is a gradient coefficient of phase field.

It is assumed that the density, special heat and conductivity coefficient are constants, and equal solid and liquid thermal conductivities are assumed, then the governing equation of temperature could be given as

\[
\frac{\partial T}{\partial t} = D_T \nabla^2 T - \left[ \frac{L_A (1-c) + cL_B}{c_p} \right] h'(\Phi) \frac{\partial \Phi}{\partial t} - \left[ \frac{(L_A - L_B)}{c_p} \right] \left[ 1 - h(\Phi) \right] \frac{\partial c}{\partial t}
\]  

Where $D_T$ is thermal diffusivity coefficient, $c_p$ is the special heat.

Eq.(11), (16) and (19) constitute the phase field model for a single-phase binary alloy.

3. Numerical methods

3.1 Initial condition and boundary condition

It is assumed that the original nucleus radius is $r_0$; the initial conditions are given as

- when $x^2 + y^2 \leq r_0^2$, \hspace{1cm}$\Phi = 1$, \hspace{1cm}$T = T_0$, \hspace{1cm}$c = c_0$  \hspace{1cm} (20)
- when $x^2 + y^2 > r_0^2$, \hspace{1cm}$\Phi = -1$, \hspace{1cm}$T = -\Delta T$, \hspace{1cm}$c = c_0$  \hspace{1cm} (21)

Where, $T_0$ is the initial temperature of undercooling molten melts, Zero-Neumann boundary condition is selected in calculations of phase field and temperature field.

3.2 Stability condition of the difference equation

It is inevitable that some truncation errors exist in the finite difference method, which is stable if the final solution changes lightly with the small fluctuations of the initial condition and boundary condition, whereas the solution is instability with the contrary condition. Harmonic analysis method is often adopted to discuss the stability condition: the error in initial value distribution is $\epsilon_{i,j}^0$, and that of next time is $\epsilon_{i,j}^{p+1}$, deduced similarly in turn for the error in $p$, $p+1$ time and so on, we can get the error increase factor $\omega(k) = \epsilon_{i,j}^{p+1} / \epsilon_{i,j}^0$. The difference equation is stable with $|\omega(k)| \leq 1$, and is unstable with $|\omega(k)| > 1$. At the same time, $\Delta t$ must be taken as the following:
\[ \Delta t \leq \min \left\{ \frac{1}{2a} \left( \frac{1}{\Delta x_{i,j,k}^2} + \frac{1}{\Delta y_{i,j,k}^2} + \frac{1}{\Delta z_{i,j,k}^2} \right) \right\} \]  

(22)

where \( \frac{1}{\Delta x_{i,j,k}^2}, \frac{1}{\Delta y_{i,j,k}^2}, \frac{1}{\Delta z_{i,j,k}^2} \) is the space step of the discrete unit.

In this paper, the even grid of space step (\( \Delta x = \Delta y \)) is adopted and the time step is \( \Delta t = \frac{\tau \Delta x^2}{5e^2} \).

The coefficient \( \frac{e^2 \Delta t}{\tau \Delta x^2} \) is maintained to be positive to assure the stability.

3.3 Program flow chart

Adopting Visual FORTRAN 6.0, 2-D dendrite growth code of the aluminum alloy has been developed, and post-process program adopted Visual C++ 6.0 were also be carried out to display

**Figure 3.** Flow chart of the dendrite growth program
dendrite growth process dynamically. Systematic environmental is P-IV 2.4G, memory is 1 G, hard disc is 80 G, and operating system is Win2000.

The flow chart of the dendrite growth program is shown in Fig.3.

Figure 4. Simulated dendrite growth process of Al alloy at different time steps
4. Simulation for dendrite growth process of Al-Si alloy

The molten metal is in thermodynamic instability during solidification; nuclei can nucleate and grow due to the crystallization driving force. As the crystal grows to a certain size, the solidified interface would become unstable under the solute fluctuation and heat disturbance, and once branches of the dendrite would be formed. Similarly, the once branches interface would also become unstable during their growth process and resulted in the secondary branches. Analogously, the third branches even higher branches could appear. So, the dendrite could have complicated side branches, which represent the most complex interface evolution pattern during solidification.

Fig4 presents the dendrite morphology evolution during its growth process. Here, $t$ is the time step. The blue and red color respectively represents the liquid metal and the solidified part. It is assumed that there has been embedded a circinal nuclei in the liquid matrix and its radius is bigger than $r_k$.

As shown in Fig4, at $t=100$, it is still a circle nuclei, and at $t=500$, it grows bigger and begins to transform to rectangle. From $t=2000$ to $t=3000$, the four corners of the rectangle grow along their tip directions and thus form the four dendrite trunks. Till $t=5000$, some little secondary side branches appear and the trunks change thinner, which is because that the solid phase in the secondary dendrite come from its corresponding trunk. During the following solidification process till $t=45000$, more and more secondary dendrites appear and grow up, at the same time, the dendrite trunk become thinner and thinner, the dendrite tip radius become smaller and smaller. At $t=55000, 65000, 75000$, some third side branches begin to appear and grow up gradually from the secondary side branches.

Therefore, it can be concluded that, during solidification process, the initial circle nuclei transforms to the rectangle one firstly, and then its corners develop to the four trunks and from which the secondary side branches generated and even the third side branches produced from secondary ones. It seems that the dendrite tip radius decrease quickly at the initial stage and change slowly at the late stage, which is mainly due to more and more side branches appear and grow up. Similarly, the dendrite grows quickly initially and gradually changes slowly even as if unchangeable finally.

5. Verification of microstructure simulation with investigations by others

Fig.5 presents the comparison of microstructure between simulated results and investigations of experimental or simulated by others with different methods.

From above results, we can see that the dendrite morphology is similar in trunks and arms growth, so the developed phase field program is accurate. Of course, this study is still at the stage of theoretical exploration, and it is necessary to be linked with engineering application further.
6. Conclusions

1. The phase field model, solution algorithm and the program of dendrite growth are introduced. The definition of initial condition, boundary condition and the stability condition of differential format are all included.

2. The dendrite morphology evolution: the initial circle nuclei transforms to the rectangle one firstly, and then its corners develop to the four trunks and from which the secondary side branches generated and even the third side branches produced from secondary ones. The dendrite tip radius decrease quickly at the initial stage and change slowly at the late stage, which is mainly due to more and more side branches appear and grow up.

3. The comparisons of dendrite morphology between simulated results and investigations by others are also presented, it is proved that the dendrite morphology are similar in trunks and arms growth, so the developed phase field program is accurate. Of course, this study is still at the stage of theoretical exploration, and it is necessary to be linked with engineering application further.

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