Effect of fluid flow on solidified equiaxed dendrite morphology evolution based on phase field - lattice Boltzmann method

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Abstract: Fluid flow can significantly change the evolution of microstructural morphology. However, relatively little is known how the fluid flow, concentration and microstructure affect each other quantitatively, which is essential to optimize processing parameters. A quantitative simulation study of Al-Cu solidified equiaxed dendrite evolution under forced flow based on phase field - lattice Boltzmann method (PF-LBM) is carried out. Results obtained are validated by Gibbs-Thomson relation at the dendrite tip. Compared with the equiaxed dendrite evolution without flow, the upstream dendrite arm is enhanced while the downstream arm is inhibited. Besides, as the inlet flow rate increases, the secondary arms attached onto the upstream primary arm and the upstream side of the primary arm normal to the inflow velocity has been well developed. Results show that sidewise instabilities of the primary dendrite arm and onset of secondary arm is caused by the local concentration perturbation and will be enhanced or inhibited by the flow. It is believed that the coupled PF-LBM method is able to handle dendrite evolution under forced flow quantitatively, which helps in investigating the solidified dendrite morphology evolution.

Key words: phase field; lattice Boltzmann method; forced flow; instability

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Solidification is one of the most universal phenomena that occurs in numerous processing procedures, such as casting, welding and additive manufacturing^[1]. It is widely accepted that the solidified microstructure, which is predominated by dendrites, mainly determines the performance of final components^[2]. Predicting microstructure evolution by simulations with processing parameters involved, especially those in which complicated heat and solute diffusion and fluid flow are coupled, has become a promising method for optimization of processing parameters to obtain a desired microstructure.

Earlier studies were mainly focused on heat or solute diffusional phase transformations^[3-5]. However, convection caused by natural or forced flow, which is inevitable in real situations and frequently plays a significant role in microstructure evolution, has been

ignored due to its complexity. Some experimental studies^[6, 7] have been conducted to investigate how the flow influences the morphology evolution. Badillo et al.^[6] conducted experiments about the equiaxed crystals of the transparent model alloy succinonitrile-acetone which developed in an undercooled melt. It was found that the upstream dendrite tip growth velocity increases, while the average of six dendrites' tip growth velocity keeps constant under forced flow. Shevchenko et al.^[7] investigated the directional solidification of Ga-25wt.% In alloy under natural flow by means of X-ray radiography. Lighter solute In was rejected from the solid into the liquid, and the buoyance-driven convection, i.e. natural convection, was produced, which in turn significantly affected the microstructure. However, much less numerical research^[8-11] has been done due to limited development of the computational method with both the microstructure evolution and the fluid flow dynamics involved. Sun et al.^[8] developed a two-dimensional (2D) cellular automation (CA) - lattice Boltzmann method (LBM) to simulate the dendritic growth of binary alloys under forced flow. It was demonstrated that the dendritic growth was strongly

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influenced by convection. Similar studies have also been done by Takaki et al.^[9, 10], wherein phase field (PF) method^[12] was employed to simulate the microstructure evolution. Qi et al.^[11] developed a PF-Naiver-Stockers (NS) method to investigate the motion of equiaxed dendritic crystals under natural and forced flow.

In order to model dendrite evolution under flow, the mathematical model should be composed of two aspects. One is for describing the solid-liquid (S-L) phase transformation, and the other is for fluid flow. For S-L phase transformations, the PF method has become the most popular method to simulate microstructure evolution, owing that explicit interface tracking is avoided, which shows exceptional advantages especially for three-dimensional dendrite evolution with complicated topological relations. In the PF method, a new parameter called order parameter ϕ is introduced to specify different phases, as in the liquid phase $\phi=-1$ and the solid phase $\phi=1$. At the S-L interface, ϕ is interpolated by the interpolation function, which is constructed delicately, and varied smoothly but sharply. The PF model can be reduced as Gibbs-Thomson relation by asymptotic matched analysis^[13]. As for the fluid flow calculation, there are mainly two available methods – the NS solver^[11] and the LBM^[9]. At mesoscopic scale, the LBM shows advantages over the NS equations. It has been shown that the fluid flow calculations using a NS solver no longer converged when the solid volume fraction went above 0.3^[8]. However, instead of simulating the transport equations (heat, mass and momentum) in a continuum way (the NS equations) or considering every particle (molecular dynamics), a distribution function which is used to describe characters of a collection of particles is utilized in the LBM. The transport of distribution function can be considered as collision and then streaming. The LBM is also a robust method and can be transformed into NS solver when the parameters are chosen properly.

In this work, PF-LBM was employed to quantitatively simulate the dendrite evolution under forced flow. Taking the Al-Cu alloy as an example, the model is quantitatively validated by Gibbs-Thomson equation at the dendrite tip. The dendrite growth and stability are also discussed.

1 Method

1.1 Phase field model

The quantitative PF model was proposed by Echebarria et al.^[5], which was originally intended to be employed in simulations of directional solidification. In order to simulate equiaxed dendrite evolution, the driving force term was rewritten, in which the directional undercooling was substituted by a unified undercooling.

The 2D governing equations for order parameter ϕ and solute c with the undercooling ΔT_0 (>0) are:

$$\tau(\vec{n}) \frac{d\phi}{dt} = \nabla \cdot (W(\vec{n})^2 \nabla \phi) + \frac{\partial}{\partial x} \left(|\nabla \phi|^2 W(\vec{n}) \frac{\partial W(\vec{n})}{\partial \phi_x} \right) + \frac{\partial}{\partial y} \left(|\nabla \phi|^2 W(\vec{n}) \frac{\partial W(\vec{n})}{\partial \phi_y} \right) - f'(\phi) + \lambda g'(\phi) \left(\frac{\Delta T_0}{m_1 c_1^0 (k-1)} - \frac{e^u - 1}{1-k} \right) \quad (1)$$

$$\frac{dc}{dt} = \nabla \cdot (Dq(\phi)c\nabla u) + \nabla \cdot j_{AT} + \nabla j_N \quad (2)$$

where $\vec{n}$ is the unit crystal orientation; $\phi_{x(y)}$ is the derivative of ϕ with respect to x or y ; m_1 , c_1^0 and k are the liquidus slope, initial liquid solute concentration and partition coefficient, respectively; D is the diffusion coefficient in the liquid;

$\nabla = \left(\frac{\partial}{\partial x}, \frac{\partial}{\partial y} \right)$ is the 2D nabla operator. $\tau(\vec{n}) = \tau_0 a^2(\vec{n})$ is the relaxation time, where $a(\vec{n}) = 1 + \varepsilon_4 \left[\frac{\phi_x^2 + \phi_y^2}{(\phi_x^2 + \phi_y^2)^2} - 3 \right]$ is

introduced to represent the crystalline anisotropy, ε_4 is the anisotropic strength, $\tau_0 = a_2(\lambda W_0^2)/D$ is the unit of relaxation time and $a_2 = 0.6267$ is constant. $W(\vec{n}) = W_0 a(\vec{n})$ is the interface width. $f(\phi) = -\frac{1}{2}\phi^2 + \frac{1}{3}\phi^3$ and $g(\phi) = \phi - \frac{2}{3}\phi^3 + \frac{1}{5}\phi^5$ are the interpolation functions associated with the double-well potential and the bulk free energy, respectively, and $f'(\phi)$, $g'(\phi)$ are the derivatives. $q(\phi) = (1-\phi)/2$ is also the interpolation function to interpolate the diffusion coefficient from the solid to liquid. $\lambda = a_1 W_0/d$ is a coupling parameter to rescale the energy caused by the solute, where $a_1 = 5\sqrt{2}/8$ is a constant and d is the chemical capillary length. j_{AT} and j_N are the anti-solute trapping^[14] and noise perturbation terms^[15], respectively. The dimensionless concentration u_i is the departure of concentration from equilibrium. Further details can be found in Ref.^[12].

1.2 Lattice Boltzmann method

The motion and rotation of dendrites were ignored. Therefore, $d\phi/dt$ in the left hand of Eq. (1) reduces to $\partial\phi/\partial t$. However, the solute in the liquid is influenced by the fluid flow, and thus dc/dt in the left hand of Eq. (2) expands into $\partial c/\partial t + U \cdot \nabla c$, where U is the local velocity of the fluid. For the solid, $U=0$ is obtained and dc/dt reduces to $\partial c/\partial t$, which is consistent with the real physics process. When the liquid runs into the solid, bounce boundary is employed. To avoid explicit tracking the S-L interface, Beckermann et al.^[16] added an additional discrete force $G_i(x,t)$ in the fluid flow equations. $G_i(x,t)$ is obtained by analogizing to the fluid flow past porous materials and the reaction force is put back into the liquid. Therefore, the LBM equation^[9] with BGK (Bhatnagar-Gross-Krook) approximation and $G_i(x,t)$ is

$$f_i(\mathbf{x} + \mathbf{c}_i \delta t, t + \delta t) = f_i(\mathbf{x}, t) - \frac{\delta t}{\tau_i} [f_i(\mathbf{x}, t) - f_i^{eq}(\mathbf{x}, t)] + G_i(\mathbf{x}, t) \delta t$$

where $f_i(x,t)$ is the distribution function in the i th direction from a statistical physics point of view, $f_i^{eq}(x,t)$ is the equilibrium distribution function, $\mathbf{c}_i$ is the velocity in the i th direction, and t , δt and τ_i are the time, time interval and relaxation time for LBM, respectively. It should be pointed out that the relaxation time τ_i in LBM is not the same as that in PF and is related to the fluid viscosity ν ,

$$\nu = \frac{1}{3} \left(\frac{\delta x}{\delta t} \right)^2 \left(\tau_i - \frac{\delta t}{2} \right) \quad (3)$$

The fluid density ρ and flow velocity U are able to be obtained by

$$\rho = \sum_{i=0}^{Q-1} f_i \quad (4)$$

$$\rho \mathbf{U} = \sum_{i=0}^{Q-1} c_i f_i \quad (5)$$

where Q is the number of discrete velocity. In this study, a 2D-nine-velocity (D2Q9) lattice arrangement is employed. The nine discrete velocities c_i are given by

$$c_i = \begin{cases} (0,0), & \text{for } i = 0 \\ c \left(\cos \left[\frac{(i-1)\pi}{2} \right], \sin \left[\frac{(i-1)\pi}{2} \right] \right), & \text{for } i = 1-4 \\ \sqrt{2}c \left(\cos \left[\frac{(2i-9)\pi}{4} \right], \sin \left[\frac{(2i-9)\pi}{4} \right] \right), & \text{for } i = 5-8 \end{cases} \quad (6)$$

where $c=\delta x/\delta t$ is the lattice velocity. The equilibrium function is given by

$$f_i^{eq}(\mathbf{x}, t) = w_i \rho \left[1 + \frac{3(\mathbf{c}_i \cdot \mathbf{U})}{c^2} + \frac{9(\mathbf{c}_i \cdot \mathbf{U})^2}{2c^4} - \frac{3\mathbf{U} \cdot \mathbf{U}}{2c^2} \right] \quad (7)$$

where w_i is the weight coefficient, which are 4/9 for $i=0$, 1/9 for $i=1-4$ and 1/36 for $i=5-8$, respectively. When additional discrete force $G_i(\mathbf{x}, t)\delta t$ is applied, an increment $\Delta \mathbf{U}$ of velocity is generated, and thus the equilibrium is perturbed^[17]. Therefore, with higher order infinitesimals neglected, the last term in Eq. (3) is

$$G_i(\mathbf{x}, t)\delta t = f_k^{eq}(\rho, \mathbf{U} + \Delta \mathbf{U}) - f_k^{eq}(\rho, \mathbf{U}) \\ = w_i \rho \left[3 \frac{\mathbf{c}_i \cdot \mathbf{U}}{c^2} + 9 \frac{(\mathbf{c}_i \cdot \mathbf{U})\mathbf{c}_i}{c^4} \right] \cdot \Delta \mathbf{U} \quad (8)$$

According to Ref.^[16], $\Delta \mathbf{U}$ is given by

$$\Delta \mathbf{U} = -\frac{2vg}{W_0^2} \left(\frac{1+\phi}{2} \right)^2 \mathbf{U} \delta t$$

To avoid complicated programming, δx and δt in the LBM were the same as the unified grid spacing dx and time interval dt in the PF method, respectively. In other words, the grid for the LBM was anchored to that for the PF method.

2 Results and discussions

The alloy employed in this study is Al-4wt.%Cu. Therefore, the initial solute concentration in the liquid c_l^0 was 0.04. Physical parameters used are listed in Table 1^[11]. The anisotropic strength used here was 0.03, while the real one is around 0.01. It should be pointed out that our main concern is not to simulate the dendrite precisely but to discover how the fluid flow influences the microstructure evolution. The artificial magnified anisotropic strength enhances the side branching, accelerates the simulation speed and does not influence the underlying mechanisms within limited computational domain.

The PF-LBM was solved in a dimensionless form, in which the spatial length was in unit of interface width W_0 and the temporal length was in unit of relaxation time τ_0 for the PF method. The ratio of W_0 to chemical capillary length $d=\Gamma/(m_l(k-1)c_l^0)=2.68 \times 10^{-2} \mu\text{m}$ was set as 5, i.e., $W_0=5d=1.342 \times 10^{-1} \mu\text{m}$. The adiabatic boundary was applied longitudinally while periodic boundary was applied vertically for the PF model. For the

Table 1: Physical parameters of Al-Cu alloy^[11]

Parameters	Value
Liquidus slope, m_l [K·(wt.%) ⁻¹]	-260
Liquidus diffusion coefficient, D [m ² ·s ⁻¹]	3.0×10^{-9}
Partition coefficient, k	0.14
Gibbs-Thomson coefficient, Γ [m·K]	2.4×10^{-7}
Anisotropic strength, ε_4	0.03
Fluid viscosity, ν [m ² ·s ⁻¹]	5×10^{-7}

LBM, to generate forced flow, a unified constant flow rate was introduced on the left boundary, which could be regarded as the inlet. Free boundary condition was adopted on the right side, and bounce boundary condition on both the top and bottom sides for the LBM. Letting $dx=W_0$ and $dt=(dx^2)/5D=4.8 \times 10^{-2} \mu\text{s}$, the scales used for length and time are in unit of dx and dt , respectively, without further statements. All cases were carried out with the simulation size of $1,024 \times 1,024$ and simulation time of 30,000 steps. A nucleation seed was put at the center of the simulation domain with a constant undercooling $\Delta T_0 = 5$ K. In order to investigate the influence of forced flow, five inlet flow rates were set to be 0.00 (without forced flow), 0.05, 0.10, 0.15, and 0.20 in the unit of dx/dt , respectively.

2.1 Validation

The results of the simulation were validated by Gibbs-Thomson equation at the dendrite tip. To quantitatively evaluate the derivation of the calculated results from analytical results, a new dimensionless variable $\Delta_{GTerror}$ is defined^[18], i.e.

$$\Delta_{GTerror} = \frac{(1-15\varepsilon_4)\Gamma\kappa - m_l(c_l^* - c_l^0)}{\Delta T_0} - 1 \quad (9)$$

where κ is the curvature and c_l^* is the solute concentration on the liquid side at the dendrite tip. c_l^* is deduced from c_s^*/k , where c_s^* is the solute concentration in the solid side. Using this approach, the concentration in the liquid is much more precise. The dendrite tip curvature is fitted by the contour of $\Phi=0$. There is no map relation for S-L interface between the sharp-interface model and PF model at the $\Phi=0$ contour. Therefore, some fluctuations might occur and the simulation is reliable if the deviations are smaller than the upper limit.

The calculated results were extracted at the last time step for all the cases. The variation of $\Delta_{GTerror}$ is shown in Fig. 1. The derivation varies around -3%, demonstrating that high accuracy of the simulation was obtained. It is unambiguous to announce that the PF results are correct within limited deviation.

2.2 Quantitative analysis for influence of fluid

The morphology evolution of equiaxed dendrite for several selected time steps is given in Fig. 2, with the upper panel showing the case of inlet flow rate equal to 0.20 and the lower panel showing the case of inlet flow rate equal to 0.00. To show the difference clearly, only half of the symmetrical dendrite is presented. It can be seen clearly that the flow significantly

Fig. 1: Variation of ΔG_{Terror} as a function of inlet flow rate

Fig. 2: Dendrite evolution at time step of 6×10^3 (purple line), 1.2×10^4 (orange line), 1.8×10^4 (green line), 2.4×10^4 (red line) and 3.0×10^4 (blue line) for inlet flow rate 0.20 (upper panel) and 0.00 (lower panel)

influences the morphology evolution, leaving the upstream dendrite arm enhanced and the downstream arm inhibited. The arms normal to the inlet flow tilt, while their lengths remain about the same as that in the lower panel. The qualitative analytical results are consistent with experiments observations [6].

In order to quantitatively describe the influence of the flow, the tip growth rates of dendrite arms were compared in two cases, one with the inlet flow rate equal to 0.00 and the other with an inlet flow rate equal to 0.02. The results are shown in Fig. 3. In the case where force flow is absent, due to fourfold symmetry of the dendrite, the tip growth rate of one dendrite arm was extracted (the orange line in Fig. 3), reaching around $v_N = 0.56 \times 10^{-2}$ at stage where stable growth has achieved. For the case in which the inlet flow rate is equal to 0.02, four different dendrite arm growth rates were obtained. For the left arm (the red line in Fig. 3), i.e. the upstream arm, stable growth rate increases to $v_L = 1.60 \times 10^{-2}$, while for the right arm (the green line in Fig. 3), i.e. the downstream arm, stable growth rate could not be obtained and decreases continuously, the reason for which will be discussed later. For the top ($v_T = 0.59 \times 10^{-2}$, the blue line in Fig. 3) and bottom arm ($v_B = 0.60 \times 10^{-2}$, the purple line in Fig.

Fig. 3: Dendrite tip growth rates. The growth rate in the case without flow is presented by orange line. The left, right, top and bottom dendrite growth rates under forced flow with flow rate equal to 0.02 are denoted by red, green, blue and purple lines, separately

3) those are normal to the inlet flow velocity, stable growth rates are nearly identical with v_N .

Besides the growth rate of the dendrite tip which only reflects one-dimensional evolution, curvature radiuses and concentrations at primary dendrites were also analyzed to reveal 2D evolution of dendrites. As can be seen from Fig. 2, the evolution tendency of the tip radius is similar with that of the growth rate. For the sake of brevity, the evolution for tip radius as a function of time step is not displayed here. Instead, the quantitative information about the radii and concentrations at a particular time step of 30,000 was compared. The radius and dimensionless concentration at the tip of primary dendrite in the case without flow are 5.13 and 1.456, respectively. In the case with inlet flow rate equal to 0.02, the curvature radiuses are 3.203 (left), 6.019 (right), 5.295 (top) and 5.085 (bottom), and dimensionless concentrations are 1.446 (left), 1.465 (right), 1.454 (top) and 1.454 (bottom). Therefore, it can be concluded that the upstream tip curvature becomes sharper while the downstream becomes blunter. This means that the upstream dendrite is less affected by the concentrated solute transported from the neighbor solid phase as a result of the asymmetrical concentration distribution, which will be discussed later.

The difference in dendrite morphologies can be explained in terms of constitutional undercooling which is resulted from fluid flow. In the case without flow [Fig. 4(a)], both the dendrite morphology and the concentration distribution show fourfold-symmetry. In the case with the inlet flow rate of 0.02 [Fig. 4(b)], in the direction of upstream, the rejected solute is transported away by the fluid. Therefore, the departure of concentration u in Eq. (1) at the dendrite tip decreases, i.e., the driving force increases. As a result, the growth of the upstream arm accelerates. In the direction of downstream, vortexes form and the rejected solute is mainly trapped in areas where the vortexes exist. As the rejected solute accumulates, the downstream arm growth rate decreases continuously. The top and bottom arms are normal to the inlet flow, so the concentration distribution at

the dendrite tips does not change severely, leaving the growth rates of the top and bottom arms almost unchanged compared with v_N .

2.3 Instability of secondary arm

It can be seen from Fig. 4(b) that the secondary arms also appeared. The secondary arms attached to the upstream primary arm and the upstream side of the primary arm normal to the inflow velocity are well developed. Similar phenomena were also observed in cases with other inlet flow rates (Fig. 5), showing that secondary arms become more and more obvious as the inlet flow rate increases. According to the classical Mullin-Sekerka theory^[19], the instability of the dendrite arms and the onset of secondary arms are caused by the local concentration perturbation and further enhanced by the force flow. Two aspects should be included, i.e., (a) the onset and (b) the development of secondary arms. (a) When local perturbations occur, secondary arm “buds” form. The perturbations are represented by the noise term j_N in Eq. (2). (b) However, when the “buds” grow into areas with high concentration of rejected solute, the local

driving force decreases, and therefore the growth rate slows down. In this situation, it is possible that the “buds” advance at the same speed as their solidified surroundings and the stability of the dendrite arm is still preserved. When local fluid flow exists, the local solute is diluted or concentrated depending on the inflow and outflow concentration of solute. For the “buds” under the red arrow 1 in Fig. 4(b), the dilute inflow decreases the concentration of the rejected solute, i.e., local driving force increases, and therefore, the “buds” are more likely to be well developed. For the “buds” under the red arrow 2, although the inflow is concentrated, the concentrated solution is then transported away by the fluid to downstream. In other words, the net increase is positive, i.e., local driving force still increases. For the “buds” under the red arrow 3 and arrow 4, due to the formation of vortices, the inflow and outflow are both concentrated solution and the concentrated solute is restricted in the vortices. Therefore, the “buds” are unlikely to develop. As illustrated in Fig. 5, the above deduction is also consistent with the instability trend as the inflow rate increases, i.e., as the inflow rate increases, the secondary arms developed mature.

Fig. 4: Dendrite morphology (top panel) and concentration distribution (bottom panel) at time step of 3×10^4 for the case without flow (a) and the case with inlet flow rate 0.02 (b), the velocity vectors of the liquid are also presented. The red arrows in (b) denote the general direction of flow.

3 Conclusions

To investigate the influence of forced flow on the dendrite evolution, a study based on phase field – lattice Boltzmann method was employed to simulate equiaxed dendrite evolution. Five cases with different inlet flow rates (0.00, 0.05, 0.10, 0.15 and 0.20) were considered. The model was validated by the Gibbs-Thomson relation at the dendrite tip, showing that high accuracy was obtained. Consistent with the previous experiment results, we found that the upstream primary dendrite arm is enhanced while the downstream arm is inhibited by the forced flow. Based on quantitative analysis, it is found that the solute rejected from the solid can be transported away by the fluid flow. The underlying cause for the instability of primary dendrite arms and onset of secondary arms is also discussed. The solution can be diluted or concentrated by the flow, which affects the driving

Fig. 5: Eventual dendrite morphologies in cases with inlet flow rate of 0.00 (purple line), 0.05 (orange line), 0.10 (green line), 0.15 (red line) and 0.20 (blue line), respectively

force of solidification and eventually leads to the difference in the development of secondary arms. The coupled PF-LBM method provides a quantitative approach to handle dendrite evolution under forced flow, which will promote studies of dendrite morphology evolution under wider conditions.

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