Modelling of dendritic growth during alloy solidification under natural convection

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Abstract

A two-dimensional (2D) lattice Boltzmann method (LBM)-cellular automaton model is presented to investigate the dendritic growth of binary alloys in the presence of natural convection. The kinetic-based LBM is adopted to calculate the transport phenomena by the evolution of distribution functions of moving pseudo-particles. To numerically solve natural convection thermal and solute transport simultaneously, three sets of distribution functions are employed in conjunction with the lattice Bhatnagar–Gross–Krook scheme. Based on the LBM calculated local temperature and concentration at the solid/liquid interface, the kinetics of dendritic growth is determined according to a local solute equilibrium approach. Thus, the physics of a complete time-dependent interaction of natural convection, thermal and solutal transport, and dendritic growth during alloy solidification is embedded in the model. Model validation is performed by comparing the simulated results with literature data and analytical predictions. The model is applied to simulate dendritic growth in binary alloys under the influence of natural convection. The effects of Rayleigh numbers and initial undercooling on dendrite growth are investigated. The results show that natural buoyancy flow, induced by thermal and solutal gradients under gravity, transports the heat and solute from the lower region to the upper region. The dendritic growth is thus accelerated in the downward direction, whereas it is inhibited in the upward direction, yielding asymmetrical dendrite patterns. Increasing the Rayleigh number and undercooling will enhance and reduce, respectively, the influence of natural flow on the dendritic growth.

Keywords: solidification, dendritic growth, natural convection, lattice Boltzmann method, cellular automaton

(Some figures may appear in colour only in the online journal)
1. Introduction

Dendrites are among the most common microstructures in metals and alloys at the end of solidification. Dendrite shapes, sizes and chemical characteristics have an important impact on the properties of the final products. Solidification is known to be always accompanied by fluid motion, created either by external forces or by natural buoyancy. In particular, natural convection is an inevitable phenomenon under gravity, due to the fact that the released latent heat and solutes lead to variations in melt density, and hence melt dynamics. Melt convection may alter the heat and solute transfer, and can thereby significantly influence the formation of dendritic patterns and solute segregation [1].

Numerous efforts have so far been devoted to characterize dendritic growth behaviour in the presence of convection using numerical models, such as phase field (PF) [2–7], front tracking (FT) [8], level set (LS) [9] and cellular automaton (CA) [10–12] methods coupled with Navier–Stokes (NS) solvers. These studies have significantly improved our understanding of the interaction between dendritic growth and melt convection. In these approaches, the melt convection is calculated by solving the NS equation and the continuity equation using the finite difference method (FDM) or finite element method (FEM). Since NS solvers are continuum-based approaches, when simulating multi-dendritic growth with melt convection, the fluid flow calculation may become difficult to converge as the solid fraction increases. In our previous work of using a NS solver [11], we found that when the solid fraction reaches about 0.3 the simulation encounters convergence problems of the fluid flow calculation. This demerit of the NS solvers makes it virtually difficult to perform micro-scale simulations of multi-dendritic growth in the presence of melt convection until the end of solidification.

Over the last decades, the lattice Boltzmann method (LBM) has rapidly developed as an attractive approach for numerically solving fluid flow [13–17]. In contrast from conventional NS solvers that are based on macroscopic continuum equations, LBM is a mesoscopic kinetic-based approach that considers flows to be composed of a collection of pseudo-particles, which are represented by a distribution function. The motion, collision and redistribution of these quasi-particles characterize the macroscopic flow dynamics in the system [13–15]. LBM can also be modified to solve energy and species transport problems due to convection and diffusion. Compared with the classic computational fluid dynamics methods, LBM offers significant advantages. First, since the LBM does not imply the assumption of a continuum fluid medium, and non-slip boundary conditions can be implemented through simple local boundary rules, it has great potential in solving fluid flow in complex geometries. Second, the main part of the algorithm is its local-type nature, rendering it well suited for parallel computation [18]. Moreover, LBM describes fluid motion at the level of individual distribution functions. Thus, the physics of the fluid particles can be naturally coupled with the related simulation techniques for crystal growth in a fluid flow.

Miller et al [19] and Medvedev and Kassner [20] developed the LBM-PF coupled models to simulate convective single dendritic growth in pure substances. In the models the phase transition is simulated by PF methods, while the melt flow and the heat transport are calculated by the LBM. Selzer et al [21] proposed a LBM-PF model to investigate the effect of forced fluid flow on the formation of dendrite and eutectic microstructures in two and three dimensions. The PF model is suitable to describe the phase transformation in multiphase, multicomponent and polycrystalline systems. The melt flow is solved by an LBM. However, the thermal and solutal fields are calculated by solving the conventional heat and mass transport equations, involving the additional convection terms, in which the fluid velocity is obtained from the LBM calculation. Chatterjee and Chakraborty [22] introduced a hybrid enthalpy-based LB model by coupling a modified thermal LBM with an adapted enthalpy-porosity technique.
to simulate thermal convection–diffusion transport involved in the dendrite growth in a pure substance.

The combination of LBM with CA approaches has also been suggested [18, 23–27]. Eshraghi et al [18] coupled the LBM with the CA method to simulate three-dimensional (3D) solute-driven dendrite growth. The proposed LBM-CA model is found to be able to simulate 3D diffusive dendrite growth with high computational efficiency and large-scale parallelization capability. The present authors [23] have constructed an LBM-CA model for the simulation of solutal dendritic growth of binary alloys with forced convection. The model adopts LBM to numerically solve forced fluid flow and solute transport. The kinetics of the solid/liquid (SL) interface evolution is determined using a local interface composition equilibrium approach proposed by Zhu and Stefanescu [28]. The model is able to quantitatively simulate dendritic growth during alloy solidification in the presence of forced convection. It is also demonstrated that the proposed LBM-CA based model is more numerically stable, computationally efficient, and simpler to be implemented for the simulation of phase transition problems coupled with melt convection than the conventional NS-based model. Yin et al [24] used a similar approach to simulate thermal-solutal dendrite growth in the presence of forced convection. Heat and mass transfer involving both diffusion and convection is also solved using LBM. Thereafter, the present authors [25] extended the LBM-CA model to the system including natural buoyancy to study the influence of natural convection on dendritic growth during alloy solidification.

The present work is a continuation of the previous study [25] on the simulation of dendritic growth under natural convection. A detailed model validation is provided, in which numerical tests of two benchmark problems of natural convection are compared with literature results. A comparison is also made between the simulations and the analytical predictions for the single dendritic growth in the presence of natural convection. The effects of natural convection on dendritic growth are discussed.

2. Governing equations and numerical algorithm

2.1. Model description

The dendritic growth of binary alloys in the presence of natural convection is considered to take place in the regime of a low Peclet number and a low Reynolds number. Externally forced convection is neglected. As the dendrite grows, latent heat and solute are released at the SL interface, which results in thermal and solutal gradients ahead of the interface. The thermal and solutal gradients yield not only thermal and solutal transport, but also natural convection under gravity. In the presence of melt convection, the thermal and solute transport is controlled by both diffusion and convection. The diffusive and convective transport of heat and solutes will result in modified temperature and composition fields, which determine the kinetics of dendritic growth. The growing dendrite, assumed to be rigid and stationary, in turn triggers an increasing complex fluid flow. It also continuously influences the thermal and solutal fields by releasing latent heat and solute. This interaction of natural convection, thermal and solutal transport, and dendritic growth continues until the end of solidification.

The basic features of the coupled model are as follows. The natural convection of the melt during dendritic growth is simulated by LBM using the standard lattice Bhatnagar–Gross–Krook (LBGK) scheme [29, 30]. The LB equation for convection calculation involves a force term that is determined by the thermal and solutal gradients in the domain. The diffusive and convective transport of solute and heat are computed by the multiple distribution function (MDF) approach of LBM, which is similar to that used in [31]. In addition, the source terms are introduced in the MDFs of the heat and solute transport to account for the released latent
heat and solute partition during dendrite growth. The MDFs of the heat and solute transport are also solved by the LBGK scheme. Based on the local temperature and composition calculated from the LBM, the kinetics of dendritic growth is determined by a local solutal equilibrium approach [28].

2.2. LBM for solving natural convection, solute and heat transport

The LBM, originating from gas kinetics theory, describes fluid flow in terms of moving and interacting individual particle distribution functions. In this work, the standard LBGK approach with the single-relaxation-time scheme [29–31] is adopted to solve the LB equations. According to the LBGK approach, the evolution equation of the particle distribution function can be expressed as

\[ f_i(x + e_i \Delta t, t + \Delta t) - f_i(x, t) = - \left[ f_i(x, t) - f_i^{eq}(x, t) \right] / \tau + F_i(x, t), \]  

(1)

where \( f_i(x, t) \) is the particle distribution function (PDF) representing the probability of finding a pseudo fluid particle in \( i \)th direction on the discrete lattice at position \( x \) and time \( t \), \( e_i \) is the discrete moving velocity of the pseudo fluid particle, \( \Delta t \) is the time step, \( \tau \) is the relaxation time, \( f_i^{eq}(x, t) \) is the equilibrium distribution function and \( F_i(x, t) \) is the force term. In the present work, \( F_i(x, t) \) is considered to be caused by the buoyancy effect due to the temperature and concentration gradients under gravity.

The D2Q9 topology [30], in which the two-dimensional (2D) space is discretized into a regular square lattice including nine velocities, is employed. The discrete velocities \( e_i \) are given by

\[
\begin{align*}
  e_i &= \begin{cases} 
  (0, 0) & i = 0, \\
  (\cos[(i - 1)\pi/2], \sin[(i - 1)\pi/2])c & i = 1 - 4, \\
  (\cos[(2i - 9)\pi/4], \sin[(2i - 9)\pi/4])\sqrt{2}c & i = 5 - 8,
\end{cases}
\end{align*}
\]

(2)

where \( c = \Delta x / \Delta t \) is the lattice speed and \( \Delta x \) is the lattice spacing. In the D2Q9 scheme, the equilibrium distribution function, \( f_i^{eq}(x, t) \), and the force term, \( F_i(x, t) \), in equation (1) can be expressed as [30, 31]

\[
\begin{align*}
  f_i^{eq}(x, t) &= w_i \rho \left[ 1 + 3 \frac{(e_i \cdot u)}{c^2} + 4.5 \frac{(e_i \cdot u)^2}{c^4} - 1.5 \frac{u^2}{c^2} \right], \\
  F_i(x, t) &= \left( 1 - \frac{1}{2\tau} \right) w_i \left[ 3 \frac{e_i - u}{c^2} + 9 \frac{e_i \cdot u}{c^4} \cdot e_i \right] \cdot F \Delta t,
\end{align*}
\]

(3)

(4)

where \( w_i \) are the weight coefficients given by \( w_0 = 4/9, w_{1-4} = 1/9 \), and \( w_{5-8} = 1/36 \), \( u \) is the macroscopic velocity of melt flow and \( F \) is the buoyancy force. The macroscopic variables such as fluid density, \( \rho \), and first momentum, \( \rho u \), can be computed from the particle distribution function by [32]

\[
\begin{align*}
  \rho &= \sum_{i=0}^{8} f_i, \\
  \rho u &= \sum_{i=0}^{8} e_i f_i + F \Delta t/2.
\end{align*}
\]

(5)

(6)

According to the Chapman–Enskog analysis, the NS equation can be recovered from the above described LB equations [33]. Accordingly, the kinematic viscosity \( \nu \), can be obtained by

\[
\nu = c^2 \Delta t (2\tau - 1)/6.
\]

(7)
Based on the Boussinesq assumption, the fluid density is a linear function of temperature and concentration, while other fluid properties are considered to be temperature and concentration independent. Thus, the buoyancy force, $F$, under gravity can be calculated by

$$F = -g\rho_0\beta_T(C_1 - C_0) - g\rho_0\beta_T(T - T_0),$$  (8)

where $g$ is the acceleration caused by gravity, $\rho_0$ is the fluid density at temperature $T_0$ and composition $C_0$, $\beta_T$ and $\beta_C$ are the solutal and thermal expansion coefficients, $C_1$ and $T$ are the local liquid composition and temperature, respectively.

In the present work, natural convection resulting from the density difference due to the thermal and solutal gradients under gravity is characterized by the dimensionless thermal Rayleigh number, $Ra_T$, and the solutal Rayleigh number, $Ra_C$, which are defined by

$$Ra_T = g\beta_T\Delta T_{Ra} L^3 / \nu \alpha,$$  (9)

$$Ra_C = g\beta_C \Delta C_{Ra} L^3 / \nu D,$$  (10)

where $L$ is the characteristic length of the computational domain, $\Delta T_{Ra}$ and $\Delta C_{Ra}$ are the maximum temperature and composition differences in liquid. In the present work, for the simulation of single dendrite growth, $\Delta T_{Ra}$ is the difference between the downward growing tip temperature in liquid, $T^*_i$, and the initial temperature, $T_0$, and $\Delta C_{Ra}$ is the difference between the downward growing tip composition in liquid, $C^*_i$, and the initial concentration, $C_0$. For the simulation of multiple dendrites, $\Delta T_{Ra}$ is the difference between the maximum temperature in liquid, $T_{l,\text{max}}$, and the initial temperature, $T_0$, and $\Delta C_{Ra}$ is the difference between the maximum concentration in liquid, $C_{l,\text{max}}$, and the initial concentration, $C_0$.

The MDF approach [32] is adopted to model the convection–diffusion of the solute and heat, by introducing two other independent distribution functions, $g_i(x, t)$ and $h_i(x, t)$. The evolution equations of the solutal and thermal distribution functions are also described by the LBGK scheme, as follows.

$$g_i(x + e_i \Delta t, t + \Delta t) - g_i(x, t) = -\left[ g_i(x, t) - g_i^{eq}(x, t) \right] / \tau_D + G_i(x, t),$$  (11)

$$h_i(x + e_i \Delta t, t + \Delta t) - h_i(x, t) = -\left[ h_i(x, t) - h_i^{eq}(x, t) \right] / \tau_a + H_i(x, t),$$  (12)

where $g_i^{eq}(x, t)$ and $h_i^{eq}(x, t)$ are the equilibrium distribution functions for concentration and temperature, $\tau_D$ and $\tau_a$ are the solutal and thermal relaxation times, $G_i(x, t)$ and $H_i(x, t)$ are the source terms due to released solute amount and latent heat during dendritic growth, respectively. The two sets of particle distribution functions in equations (11) and (12) are incorporated with the flow evolution equation of equation (1) through velocity vector, $u$, in the relevant equilibrium distribution functions, $g_i^{eq}(x, t)$ and $h_i^{eq}(x, t)$, which are defined by

$$g_i^{eq}(x, t) = w_i C \left[ 1 + 3(e_i \cdot u)/c^2 + 4.5(e_i \cdot u)^2/c^4 - 1.5u^2/c^2 \right],$$  (13)

$$h_i^{eq}(x, t) = w_i T \left[ 1 + 3(e_i \cdot u)/c^2 + 4.5(e_i \cdot u)^2/c^4 - 1.5u^2/c^2 \right].$$  (14)

The macroscopic concentration $C$ and temperature $T$ in equation (8) can be calculated as

$$C = \sum_{i=0}^{8} g_i$$  (15)

$$T = \sum_{i=0}^{8} h_i.$$  (16)

Similar to the relationship between the kinematic viscosity and the relaxation time, the solutal and thermal diffusivities, $D$ and $\alpha$, are related to the relaxation times, $\tau_D$ and $\tau_a$, by the following equations

$$D = c^2 \Delta t (2\tau_D - 1)/6 \quad \text{and} \quad \alpha = c^2 \Delta t (2\tau_a - 1)/6.$$  (17)
To calculate the unknown particle distribution functions at the boundary nodes of a 2D domain \( \{(x, y) | 0 \leq x \leq L, 0 \leq y \leq L\} \), the following boundary conditions are adopted in the present work, unless otherwise specified. For natural convection calculations, four surfaces of the domain are treated as solid walls. The non-slip boundary condition is, therefore, applied at the domain surfaces and the SL interface by imposing the bounce-back rule. For solutal transport calculation, the zero-flux boundary condition of \( \partial_x C \big|_{x=0,L} = 0 \) and \( \partial_y C \big|_{y=0,L} = 0 \) is implemented on the four surfaces of the calculation domain. Since convection vanishes in the solid and the solute diffusion coefficient in the solid phase is about three orders of magnitude smaller than that in the liquid, solid diffusion is neglected and solute transport is only computed in the liquid phase. The bounce-back scheme is thus also applied at the SL interface for solutal field calculation. The temperature at the domain boundary walls is fixed to \( T_0 \) by employing the non-equilibrium extrapolation scheme \([34]\).

### 2.3. Kinetics of dendritic growth

The kinetics of dendritic growth is determined by the solute equilibrium approach, proposed by Zhu and Stefanescu \([28]\). In this approach the evolution of the SL interface is considered to be driven by the difference between the local interface equilibrium concentration and the local actual liquid concentration. The interface equilibrium concentration, \( C_{eq}^l \), can be computed by

\[
C_{eq}^l = C_0 + [(T^* - T_{eq}^l) + \Gamma K(1 - 15\varepsilon \cos[4(\theta - \theta_0)])]/m, \tag{18}
\]

where \( T^* \) is the interface temperature, \( T_{eq}^l \) is the equilibrium liquidus temperature at the initial concentration \( C_0 \), \( \varepsilon \) is the anisotropic degree of the SL interface energy, \( m \) is the liquidus slope, \( \Gamma \) is the Gibbs–Thomson coefficient, \( K \) is the curvature of the SL interface, \( \theta \) is the growth angle between the interfacial normal direction and the horizontal direction and \( \theta_0 \) is the angle of the preferential growth direction with respect to the horizontal direction. The calculated local interface equilibrium concentration is compared with the local actual liquid concentration at the interface, \( C^*_l \), which is determined by solving the LB equations. If the concentration difference in an interface cell satisfies the condition of \( \Delta C = C_{eq}^l - C^*_l > 0 \), then the solid fraction should increase. According to the solute equilibrium condition, the increased solid fraction, \( \Delta \phi_s \), at the interface cell in one time step interval, \( \Delta t \), can be evaluated by

\[
\Delta \phi_s = \left(\frac{C_{eq}^l - C^*_l}{C_{eq}^l(1-k)}\right), \tag{19}
\]

where \( k \) is the solute partition coefficient. As the solid fraction increases, the solute is rejected at the SL interface. Solute partition between liquid and solid at the SL interface is considered according to \( C_s^*_l = kC^*_l \). The rejected solute \( \Delta C_{int} \), and the increased temperature \( \Delta T_{int} \) by the released latent heat, in an interface cell at each time step can be evaluated by

\[
\Delta C_{int} = \Delta \phi_s C^*_l (1-k) \quad \text{and} \quad \Delta T_{int} = \Delta \phi_s \Delta H/C_p, \tag{20}
\]

where \( \Delta H \) is the latent heat and \( C_p \) is the specific heat. Thus, the source terms in equations (11) and (12) can be calculated with

\[
G_i(x,t) = w_i \Delta \phi_s C^*_l (1-k) \tag{21}
\]

\[
H_i(x,t) = w_i \Delta \phi_s \Delta H/C_p. \tag{22}
\]

If at time \( t = t_n \) the sum of the solid fraction in an interface cell equals one, then the state of this cell is assigned as solid and a new interface cell is explicitly captured. The exact SL front is, however, implicitly scaled by the solid fraction within each interface cell. The local interface curvature \( K \), and the growth angle \( \theta \), in equation (18) can be calculated according to
the solid fraction gradient at the SL interface by
\[ K = \left[ 2\partial_x\varphi_s\partial_y\varphi_s\partial^2_x\varphi_s - (\partial_x\varphi_s)^2 \partial^2_y\varphi_s - (\partial_y\varphi_s)^2 \partial^2_x\varphi_s \right] \cdot \left[ (\partial_x\varphi_s)^2 + (\partial_y\varphi_s)^2 \right]^{-3/2} \] (23)

\[ \theta = \arccos \left( \frac{\partial_x\varphi_s}{\left[ (\partial_x\varphi_s)^2 + (\partial_y\varphi_s)^2 \right]^{1/2}} \right) \] (24)

Equations (23) and (24) are solved by a centred finite difference scheme with second-order accuracy to the partial derivatives of solid fraction.

2.4. Model coupling and numerical solution sequence

The coupling of natural convection, thermal and solutal transport, and dendritic growth is implemented by numerically solving the governing equations, which are described above iteratively, as follows.

1. Initialize the simulation system with domain length, grid size, initial temperature and composition, and solid seeds with preferential crystallographic orientations.
2. Choose relaxation time for fluid flow calculation as \( \tau = 2.0 \). Based on the Prandtl number \( (Pr = \frac{\nu}{\alpha}) \) and the Schmidt number \( (Sc = \frac{\nu}{D}) \), the relaxation times for the LBM calculation of thermal and solutal transport are determined according to \( \tau_a = \frac{(\tau - 0.5)}{Pr} + 0.5 \) and \( \tau_D = \frac{(\tau - 0.5)}{Sc} + 0.5 \).
3. Calculate the flow velocity, temperature and concentration fields by solving equations (1)–(17). The natural convection is triggered by the temperature and concentration gradients in the domain through equations (1), (4), (6) and (8)–(10). On the other hand, the natural convection will in turn influence the solute and heat transport by the flow velocity vector involved in equations (13) and (14).
4. Calculate the dendrite growth according to equations (18), (19), (23) and (24). The interface temperature and concentration in equations (18) and (19) are obtained by LBM calculation in step (3). During dendritic growth, the released latent heat and solute will increase the temperature and concentration at the SL interface. This phenomenon is simulated by the calculation of the source terms using equations (21) and (22), which are included in the solutal and thermal distribution functions of equations (11) and (12). Moreover, the implementation of the non-slip boundary condition at the SL interface renders the growing dendrite in a configuration where it in turn influences the evolution of flow field.
5. Update the fluid flow, temperature, concentration and solid fraction fields.
6. Proceed to the next interval of time step from step (3) until the end of simulation.

3. Results and discussion

3.1. Validation of LBM for the calculation of natural convection

To test the validity of the LBM for the modelling of natural convection, a benchmark problem of 2D thermal fluid dynamics in a square heated cavity is simulated. The square domain with a length of \( H \) is divided into 256 × 256 cells. The horizontal walls are assumed to be adiabatic. The left and right vertical walls are isothermal and imposed with a high temperature, \( T_h = 1 \), and a low temperature, \( T_c = 0 \), respectively. The Prandtl number \( Pr = \frac{\nu}{\alpha} \) is set to be 0.71.

Figure 1 shows the simulated steady-state isotherm lines with various Rayleigh numbers from \( 10^3 \) to \( 10^6 \). The thermal transfer mechanism with various Rayleigh numbers can be observed from the isotherm lines. At a low Rayleigh number \( (Ra_T = 10^3) \), the isotherm
lines are roughly parallel along the vertical direction, indicating that the thermal transfer is dominated by conduction between the hot and cold walls. As the Rayleigh number is increased, the predominant thermal transfer mechanism shifts from conduction to convection. When the Rayleigh number is increased to $Ra_T = 10^6$, the isotherm lines in the centre region become parallel along the horizontal direction and the isotherm lines keep roughly vertical only at the thin boundary layer near the cold and hot walls. The features of the isotherm lines presented in figure 1 are found to compare well with the results reported in [35].

In our previous work [36], the average Nusselt number at the hot wall, $Nu$, was computed via

$$Nu = -\frac{1}{T_h - T_c} \int_0^H \left. \frac{\partial T}{\partial x} \right|_{\text{hot wall}} dy.$$  \hspace{1cm} (25)

The calculated average Nusselt numbers at the hot wall with various Rayleigh numbers were compared with the data calculated by de Vahl Davis using the stream-function-vorticity approach [37]. It is found that the results of the LBM calculation coincide well with the literature data. The maximum relative error is within 3.0% for the cases considered.

The second case used for validation is the simulation of Rayleigh–Bernard convection in a rectangular cavity with width, $L$, and height, $H$, ($L/H = 2$). The calculation domain consists of $81 \times 41$ lattice points. The zero-flux boundary condition is adopted for both fluid flow and temperature field at the left and right walls. The top and bottom surfaces of the domain are treated as solid walls for fluid flow calculation using the bounce-back scheme. The constant temperatures at the top and bottom walls are imposed as $T_c = 0$ and $T_h = 1$, respectively, by employing a non-equilibrium extrapolation approach [34]. In the beginning of the simulation, the fluid is assumed to be static and an average temperature is taken as $T_0 = (T_h + T_c)/2$ in the domain. The Prandtl number is chosen as 0.71. After the system reaches the steady state, the average Nusselt number, $N_u$, is calculated, which is defined by [38]

$$N_u = 1 + \int_0^H \int_0^L \frac{(u_y \cdot T)}{\alpha \cdot (T_h - T_c)} dx dy.$$  \hspace{1cm} (26)

Figure 2 presents a comparison of the calculated average Nusselt number as a function of Rayleigh number with the data obtained by Clever and Busse [39] using a Galerkin approach. It can be noted that the simulated results of the present model are very close to the data in the literature.
3.2. Growth of single dendrite

Figure 3 presents the evolution of the composition, temperature, flow fields, and the morphology of a single dendrite freely growing in an undercooled melt of $\Delta T = 0.9$ K with natural convection. The calculation domain consists of $500 \times 500$ grid points with a grid size $\Delta x = 0.4 \mu m$. Since in the present work the single-relaxation-time scheme is adopted to solve the LB equations on a single grid domain, the values of relaxation times are confined to a relative narrow range for the quantitative LBM calculations. According to equations (7) and (17), the relaxation times of fluid flow, heat and mass transport are related to the Prandtl number ($Pr = n/\alpha$) and Lewis number ($Le = \alpha/D$). To increase the accuracy of the LBM calculations, non-realistic physical parameters are used, in which the kinematic viscosity, and the thermal and solutal diffusivities are set to be the same order of magnitude. Thus, the three relaxation times could be close and ensure the accuracy of LBM calculations. The physical parameters used are as follows: partition coefficient $k = 0.1$; liquidus slope $m = -2.3$ K mol$^{-1}$ mol$^{-1}$; Gibbs–Thomson coefficient $\Gamma = 6.9 \times 10^{-6}$ mK; specific heat $C_p = 1940$ J m$^{-3}$ K$^{-1}$; melting temperature of pure melt, $T_m = 331.23$ K; latent heat $\Delta H = 5 \times 10^3$ J m$^{-3}$; kinematic viscosity $v = 6.0 \times 10^{-9}$ m$^2$ s$^{-1}$; solutal diffusion coefficient $D = 1.0 \times 10^{-9}$ m$^2$ s$^{-1}$; thermal diffusion coefficient $\alpha = 3.0 \times 10^{-9}$ m$^2$ s$^{-1}$; and the degree of interface energy anisotropy $\varepsilon = 0.0267$. The initial melt composition of the binary alloy is
Tip velocity, $V$ (10^{-5}$m/s)

Time, $t$ (s)

Upstream tip
Downstream tip
Horizontal tip
Without flow

Tip temperature, $T^*$ (K)

Time, $t$ (s)

Upstream tip
Downstream tip
Horizontal tip
Without flow

(a) (b)

Tip concentration, $C_1$ (wt%)

Time, $t$ (s)

Upstream tip
Downstream tip
Horizontal tip
Without flow

(c) (d)

Tip concentration ratio, $C_{eq}/C_1$

Time, $t$ (s)

Upstream tip
Downstream tip
Horizontal tip
Without flow

Figure 4. Time histories of (a) growth velocity, (b) temperature, (c) concentration and (d) the ratio of the equilibrium concentration and actual concentration of dendrite tips for the case of figure 3.

$C_0 = 0.3$ mol%. At the beginning of the simulation, a solid seed with the composition of $kC_0$ and a preferential crystallographic orientation of $0^\circ$ with respect to the horizontal direction is placed at the centre of the domain. The Rayleigh numbers are taken as $Ra_T = Ra_C = 5 \times 10^3$. It can be seen from figure 3 that at the early stage of dendritic growth, the natural convection, which is induced by the thermal and solutal gradients under gravity, is relatively weak and flows smoothly around the dendrite from the bottom to the top. The shape of the small dendrite in its incipient growth stage is almost symmetrical. Because of the bounce-back effect from the solid boundaries, two large vortices appear on the right-hand and left-hand sides of the dendrite, respectively. As the dendrite grows further, latent heat and solute are continuously released at the SL interface, which gradually enhances the natural convection. When the flow velocity is large enough, four rotating vortices develop between the dendrite arms. The flow flux in the vicinity of the dendrite is, however, always upwards, which transports the heat and solute from the lower region to the upper region and thus produces asymmetrical thermal and solutal distributions. Since the kinetics of dendritic growth is determined by the local temperature and composition, the asymmetrical thermal and solute fields lead to an asymmetrical dendritic shape. The downward tip grows faster while the growth of the upward tip is retarded, as shown in figures 3(c) and (d).

Figure 4 presents the evolutions of the tip velocity, temperature, and composition, respectively, corresponding to the case in figure 3. The tips growing in the downward and
upward directions are referred to as the upstream tip and the downstream tip, respectively. The data of the dendritic growth in a purely diffusive environment are also plotted in the figures for comparison. As shown in figure 4(a), in the initial stage, all tip velocities begin from a large value. As the dendrite grows, the solute and latent heat is released and enriched in the SL interface, which quickly increases the composition and temperature in front of the tips. According to equations (18) and (19), the increased interface temperature, $T^*$, reduces the interface equilibrium concentration, $C^*$. Together with the increasing actual liquid concentration at the interface, $C^*_l$, the tip velocities thus fall down rapidly. After a transient period, the temperature and concentration ahead of the tips reach approximately stable values at different levels. This indicates that the heat and solute rejection is balanced by the heat and solute transport due to convection and diffusion. The steady-state temperatures of the upstream and downstream tips are lower and higher than that of pure diffusion, respectively. The horizontal tip has an approximately intermediate tip temperature that is close to the one without convection as shown in figure 4(b). On the contrary, it can be seen from figure 4(c) that the steady-state concentration of the upstream tip is higher than that of the downstream tip, due to the fact that the faster growing upstream tip releases more solute, and it is accumulated in the upstream region. Nevertheless, the difference in the equilibrium composition and the actual tip composition in liquid for the downstream tip is smaller, that is, $(C^*_{eq} - C^*_l)_{down} < (C^*_{eq} - C^*_l)_{up}$, leading to a lower steady-state growth velocity for the downstream tip compared to the upstream tip, as shown in figures 4(a) and (d).

To further investigate the effect of natural convection on dendrite growth, simulations are performed with various thermal and solutal Rayleigh numbers. The other calculation conditions are identical with those of figure 3. The simulated temperature fields, composition fields, flow fields, and dendrite shapes with various Rayleigh numbers ((a) $Ra_T = Ra_C = 0$; (b) $Ra_T = Ra_C = 2 \times 10^3$ and (c) $Ra_T = Ra_C = 8 \times 10^3$) are presented in figure 5. When the thermal and solutal Rayleigh numbers are taken as zero, the dendrite is growing in a purely diffusive environment. Symmetrical features of the concentration and temperature fields, and the dendritic shape can be obtained as shown in figure 5(a). On the other hand, when imposing non-zero Rayleigh numbers, natural convection emerges, which carries the rejected solute and latent heat away from the lower region to the upper region, resulting in asymmetrical concentration and temperature fields, and flow-specific dendritic patterns. In addition, when the Rayleigh numbers are increased from $2 \times 10^3$ to $8 \times 10^3$, the natural convection becomes stronger. Consequently, the effect of flow on the dendritic shape is enhanced, as shown in figures 5(b) and (c). For the case of Rayleigh numbers $Ra_T = Ra_C = 2 \times 10^3$, the steady-state growth velocity ratio of the upstream and downstream tips, $V_{up}/V_{down}$, is 1.826. When the Rayleigh numbers are taken as $Ra_T = Ra_C = 8 \times 10^3$, the steady-state tip velocity ratio, $V_{up}/V_{down}$, is increased to 4.588. The distances from the centre of dendrite (the location of seed) to the upstream and downstream tips are referred to as the upstream arm length, $L_{up}$, and the downstream arm length, $L_{down}$, respectively. The length ratio of the upstream and downstream arms, $L_{up}/L_{down}$, for various Rayleigh numbers is measured and compared in figure 6. It is noted that the length ratio of the upstream and downstream arms increases with the Rayleigh number.

Melt flow changes not only the dendrite shape, but also with the overall growth kinetics. Figure 7 shows the evolution of solid fraction with time for various Rayleigh numbers. Generally, as the Rayleigh numbers increase, the time required for reaching the same solid fraction is decreased. It is evident that convection is more efficient than diffusion to transport the rejected solute and heat away from the SL interface into the far field of the melt. The local solute and heat enrichment at the interface is, therefore, lower than for the case of pure
**Figure 5.** Simulated dendrite morphologies freely growing in an undercooled melt ($\Delta T = 0.9 \, K$, $C_0 = 0.3\, \text{mol}\%$) with various Rayleigh numbers of (a) $Ra_T = Ra_C = 0$, (b) $Ra_T = Ra_C = 2 \times 10^3$ and (c) $Ra_T = Ra_C = 8 \times 10^3$: the upper row showing the concentration fields and the bottom row showing the temperature fields (domain: $500 \times 500$ with $\Delta x = 0.4 \, \mu m$).

**Figure 6.** The length ratio of upstream arm and downstream arm varies with Rayleigh number.

diffusion, which accelerates the overall solidification rate. The higher Rayleigh numbers produce stronger natural convection, leading to faster solidification.

To examine the role of the initial undercooling on dendritic morphology, both with and without natural flow, simulations are performed with various initial melt undercoolings. For the case with natural convection, the thermal and solutal Rayleigh numbers are taken as $Ra_T = Ra_C = 2 \times 10^3$. The other calculation conditions are identical with those of figure 3. Figure 8 presents the simulated dendrite shapes with natural convection (the solid line), showing asymmetrical morphologies compared to those growing in the pure diffusion
Figure 7. Time histories of solid fraction with various Rayleigh numbers ($\Delta T_0 = 0.9$ K, $C_0 = 0.3$ mol%).

Figure 8. Comparisons between morphologies of the convective (solid line) and pure-diffusive (dashed line) dendrites freely growing in an undercooled melt with various initial undercoolings of (a) $\Delta T_0 = 0.5$ K ($F_s = 14.0\%$), (b) $\Delta T_0 = 0.7$ K ($F_s = 11.4\%$) and (c) $\Delta T_0 = 0.9$ K ($F_s = 10.5\%$) ($F_s$: total solid fraction; $Re=Re_T = Re_C = 2 \times 10^3$; domain: $500 \times 500$ with $\Delta x = 0.4$ $\mu$m).

The condition (see dashed line). Each case is simulated to produce an identical upstream arm length equal to 80 $\mu$m for the convective dendrite. Note that with increasing undercooling, the dendritic shape becomes thinner for both extreme cases; namely, pure diffusion and natural convection. In addition, as undercooling is increased, the asymmetrical degree of the dendrite shape with natural convection seems to be reduced. When the undercooling is 0.5 K, 0.7 K and 0.9 K, the corresponding length ratio of the upstream arm and the downstream arm, $L_{up}/L_{down}$, is 1.798, 1.584 and 1.416, respectively, indicating the decreasing effect of natural convection on dendrite growth with increased undercooling. This trend coincides with the PF-NS simulation results of the dendritic growth of pure succinonitrile (SCN) under natural convection reported by Tönhardt and Amberg [4]. They explained that the size of the dendrite becomes larger as the undercooling is reduced, which enhances the natural convection [4]. We have also considered another reason. For the case with lower undercooling, the growth velocity is slower and thereby more time is available for the heat and solute transport from the upstream region to the downstream region by convection, leading to the more pronounced asymmetrical dendrite morphology.
3.3. Comparison with the Lipton–Glicksman–Kurz analytical model

The Lipton–Glicksman–Kurz (LGK) analytical model [40] describes the steady-state growth of a branchless needle dendrite freely growing at a given melt undercooling. A constant initial composition of the melt far away from the dendrite tip is assumed. Neglecting the kinetic effect, the total undercooling, consisting of thermal, solutal and curvature contributions, for dendritic growth in two dimensions is given by

$$\Delta T = \left( \frac{\Delta H}{c_p} \right) \Omega_T + \frac{k \Delta T_0 \Omega_C}{1 - (1 - k) \Omega_C} + \frac{\Gamma}{R},$$  \hspace{1cm} (27)

where $R$ is the steady-state tip radius, $\Delta T_0 = m C_0 (1 - 1/k)$ is the equilibrium liquidus–solidus temperature interval at composition $C_0$, $\Omega_T$ is the dimensionless thermal undercooling and $\Omega_C$ is the dimensionless solutal supersaturation. For the case of purely diffusion-controlled growth, $\Omega_T$ and $\Omega_C$ are calculated from the 2D Ivantsov solutions by

$$\Omega_T = Iv \left( P_T \right) = \sqrt{\pi P_T} e^{P_T} \text{erfc} \left( \sqrt{P_T} \right),$$

$$\Omega_C = Iv \left( P_C \right) = \sqrt{\pi P_C} e^{P_C} \text{erfc} \left( \sqrt{P_C} \right),$$  \hspace{1cm} (28a, 29a)

where $P_T \equiv V R / (2 \alpha)$ and $P_C \equiv V R / (2 D)$ are the thermal and solutal Peclet numbers, $V$ is the steady-state tip growth velocity and the function $\text{erfc} (x)$ denotes $2 \pi^{-1/2} \int_x^\infty e^{-t^2} \, dt$.

The selection criterion of the dendritic tip radius in an undercooled alloy melt is obtained as

$$R = \frac{d_0}{\sigma^*} \left[ 2 P_C \left( \frac{k \Delta T_0 (\Delta H / c_p)}{1 - (1 - k) \Omega_C} \right) + P_T \right]^{-1},$$  \hspace{1cm} (30)

where $d_0 = \Gamma / (\Delta H / c_p)$ is the capillary length and $\sigma^*$ is the selection parameter. According to the solvability theory, $\sigma^*$ is a function of the anisotropy parameter $\varepsilon$ [41, 42]. In the present work, the selection parameter $\sigma^*$ is determined according to the linearized solvability theory derived by Barbieri and Langer [43]. For the case of $\varepsilon = 0.0267$ used for the simulations, the corresponding $\sigma^*$ for two dimensions calculated by the linearized solvability theory is 0.106 54 [43].

Equations (27)–(30) constitute the LGK model for the purely diffusion-controlled dendrite growth [40]. To extend the LGK model to the system including convection, the modified 2D thermal and solutal Ivantsov solutions incorporating the convective effect are derived based on the method proposed by Cantor and Vogel [44]. Thus, the dimensionless thermal undercooling, $\Omega_T$, and the dimensionless solutal supersaturation, $\Omega_C$, in the presence of convection are calculated by

$$\Omega_T = \sqrt{\pi P_T} e^{P_T} \left[ \text{erfc} \left( \sqrt{P_T} \right) - \text{erfc} \left( \sqrt{P_T} \left( 1 + 2 \delta_T / R \right) \right) \right],$$

$$\Omega_C = \sqrt{\pi P_C} e^{P_C} \left[ \text{erfc} \left( \sqrt{P_C} \right) - \text{erfc} \left( \sqrt{P_C} \left( 1 + 2 \delta_C / R \right) \right) \right],$$  \hspace{1cm} (31a, 32a)

where $\delta_T$ and $\delta_C$ are the thickness values for the thermal and solutal layers, respectively, influenced by the convection. According to the 2D convection model described in [45], the thermal and solutal boundary layer thicknesses can be expressed as

$$\frac{\delta_T}{\ell_C} = \frac{B}{A} (Ra_T)^{-1/4} \left[ 1 + \left( \frac{Pr}{Sc} \right)^{1/2} \frac{\delta_T}{N} \right]^{-1/4}$$

$$\frac{\delta_C}{\ell_C} = \frac{B}{A} (Ra_C)^{-1/4} \left[ 1 + \left( \frac{Sc / Pr}{N} \right)^{1/2} \right]^{-1/4},$$  \hspace{1cm} (33a, 34a)
where \( l_C \) is a characteristic length scale for convection, \( Le = Sc/Pr = \alpha/D \) and \( \tilde{N} \) is a buoyancy parameter given by

\[
\tilde{N} = \frac{\beta_C (C^*_T - C_0)}{\beta_T (T^*_T - T_0)}.
\]

where \( T^*_T \) and \( C^*_T \) are the temperature and solute concentration in the liquid at the downward growing dendrite tip, respectively. Based on the approach described in [46], the characteristic length scale \( l_C \) is taken as the downward growing tip radius \( R \), and the value of \( B/A \) is estimated as 2.2. Substituting the modified Ivantsov solutions of equations (31) and (32) into equations (27) and (30), the analytical predictions of the steady-state tip velocity and radius of dendrite growth with melt convection can be obtained.

The simulations of single dendritic growth in an undercooled melt of \( \Delta T = 0.9 \text{ K} \) and initial composition of \( C_0 = 0.3 \text{ mol\%} \) are performed with various thermal and solutal Rayleigh numbers. The other conditions are identical with those of figure 3. After dendritic growth reaches steady state, the velocity and radius of the downward growing tip are measured. The tip velocities are directly measured from the interface cells around the downward growing tip and the average values are calculated. The tip radii are obtained using a parabolic fitting to the simulated dendrite shape. The details of measuring steady-state tip velocity and radius can be found elsewhere [28]. Figure 9 presents a comparison between the simulations and the analytical results for the steady-state velocities and radii of the downward growing tips with various Rayleigh numbers. It can be seen that the changes in both velocity and radius with the Rayleigh number predicted by the two models show the same tendency: the tip velocity increases, while radius decreases with increasing Rayleigh numbers. The simulated growth velocities and radii are found to be slightly higher and lower than the analytical predictions, respectively. Nevertheless, the simulated data appear reasonably close to the analytical solutions.

### 3.4. Growth of multiple dendrites

The model is also applied to simulate the growth of multiple dendrites with natural convection. The physical parameters of succinonitrile-acetone (SCN-Ace) alloys [4, 40, 46] are used for this simulation: \( k = 0.1 \); \( m = -2.16 \text{ K mol\%}^{-1} \); \( \Gamma = 6.52 \times 10^{-8} \text{ mK} \); \( \Delta H = 4.62 \times 10^4 \text{ J m}^{-3} \); \( C_p = 1940 \text{ J m}^{-3} \text{ K}^{-1} \); \( T_m = 331.23 \text{ K} \); \( v = 2.6 \times 10^{-6} \text{ m}^2 \text{ s}^{-1} \); \( D = 1.27 \times 10^{-9} \text{ m}^2 \text{ s}^{-1} \).
Figure 10. Evolution of the multiple dendrites growing in an undercooled melt ($C_0 = 0.5$ mol%, $\Delta T = 1$ K and the cooling rate is 20 K s$^{-1}$) with Rayleigh numbers of $Ra_T = Ra_C = 5 \times 10^3$ (a) $F_s = 2\%$, $t = 0.06$ s, (b) $F_s = 10\%$, $t = 0.15$ s, and (c) and (d) $F_s = 18\%$, $t = 0.19$ s ($F_s$: total solid fraction): (a)–(c) showing the concentration field and (d) showing the temperature field (domain: $500 \times 500$ with $\Delta x = 1 \mu m$).

and $\alpha = 1.12 \times 10^{-7}$ m$^{-2}$ s$^{-1}$. However, to generate an enhanced natural convection field, the thermal and solutal Rayleigh numbers with $Ra_T = Ra_C = 5 \times 10^3$ are imposed, rather than using the realistic thermal and solutal expansion coefficients of SCN-Ace alloys. Initially, five crystal seeds with randomly assigned preferred growth orientations are placed on a square computational domain of a $500 \times 500$ grid with a uniform grid size $\Delta x = 1 \mu m$. The domain is filled with the undercooled melt of $\Delta T_0 = 1$ K and $C_0 = 0.5$ mol%. The cooling rate is set to be 20 K s$^{-1}$. Figure 10 presents the evolution of the composition, temperature, and flow fields, and the morphology of multiple dendrites with natural convection. It can be seen that at the early stage of the solidification, the natural flow is weak and the convective effect is less important. The small dendrites, growing along various preferential crystallographic orientations, present nearly symmetrical patterns. As the dendrites grow, the natural flow becomes gradually stronger and more complex with several developed rotating vortices. However, the flow flux in the vicinity of the dendrites is almost directly upwards. The growth of the dendritic arms in the inner region is mostly suppressed by the nearby dendrite and is not much affected by convection. On the other hand, the growth of the dendritic arms in the outer region is evidently influenced by natural convection. The dendrite arms in the lower region seem to be more developed compared to the ones in the upper region. The mechanism is identical with the case of the single dendrite growth. The upward natural flow transports the released heat and solute from the lower region to the upper region, providing a relative larger driving force for the growth of the dendrite arms in the upstream region.

4. Conclusions

An LBM-CA model is proposed and applied to simulate dendritic growth with natural convection during alloy solidification. The model calculates natural convection, thermal and solutal transport using the kinetic-based LBM. A force term is introduced into the LB equation, rendering the approach capable of simulating natural convection induced by both temperature and concentration gradients under gravity. On the other hand, the LB thermal and solutal distribution functions are incorporated with a flow velocity vector and the source terms that account for the released latent heat and solute at the SL interface. Thus, the thermal and solute transport governed by both convection and diffusion during dendrite growth can be calculated in a straightforward manner. Based on the LBM calculated local temperature and concentration, the kinetics of dendritic growth is determined according to a local solute equilibrium approach.
The validity of the LBM for the simulation of natural convection is tested by conducting simulations of two benchmark problems, including the thermal fluid dynamics in a square cavity and the Rayleigh–Bernard convection in a rectangular cavity. The predictions, namely, the steady-state isotherm lines and average Nusselt numbers with various Rayleigh numbers agree well with the data in the literature.

The model is applied to simulate single dendrite growth of binary alloys with natural convection. The results show that the natural flow, caused by the thermal and solutal gradients under gravity, transports the heat and solute from the lower region to the upper region, for the case when the solute is less dense than the solvent. It thus promotes and retards the growth of the downward tip and the upward tip, respectively, producing the asymmetrical dendrite morphology. With increasing thermal and solutal Rayleigh numbers, the growth velocity ratio of the upstream and downstream tips is increased, indicating an enhanced effect of natural flow on the dendritic growth. Natural convection is also found to accelerate the overall solidification rate. The effects of the initial undercooling on dendrite morphology are investigated. When increasing the undercooling, the dendrite shape becomes thinner for both cases with natural convection and pure diffusion. Moreover, the influence of natural convection on dendrite growth is found to be reduced as undercooling increases.

The steady-state growth velocity and radius of the upstream tip with various Rayleigh numbers are simulated and compared with the predictions of the modified LGK analytical model, which incorporates the effect of convection. The simulated data are found to be in reasonable agreement with the analytical predictions.

The model is also able to simulate the growth of multiple dendrites with various orientations in the presence of natural convection. It is observed that at the early stage of the solidification the natural flow is weak and the convective effect is less important. As the dendrites grow, the natural flow becomes gradually stronger and more complex. The flow flux in the vicinity of dendrites is, however, almost directly upwards, leading to more developed dendrite arms in the lower region than those in the upper region. Moreover, the influence of natural flow on the dendrite growth is more evident in the outer region compared to that in the inner region. The simulation results illustrate the interactive nature of buoyancy flow, thermal and solute transport, and dendrite growth during alloy solidification.

The improvement of the present model by using a multiple-relaxation-time scheme [47] remains as one of the tasks for the further development of the LBM-CA model, so that the model can be applied to quantitatively simulate the dendritic growth under natural convection of real alloys.

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