Multiphase Numerical Simulations of Binary Alloy Lamellar Eutectoid Growth

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Abstract. Based on the KKSO model\textsuperscript{[1]}, phase field and solute field are coupled to simulate the growth of lamellar pearlite in eutectoid transformation of Fe-C binary alloy. The influence of interface energy and diffusion coefficient on the microstructure and growth morphology of lamellar pearlite is studied. The results show that eutectoid lamellar morphology changed from regular symmetry to low symmetry and small amplitude oscillation, the growth rate of lamellar microstructure decreases and the morphology of ferrite phase interface is changed from horizontal to convex with the increase of interfacial energy. With the increase of diffusion coefficient, the growth rate of lamellar pearlite microstructure increases and the morphology of pearlite interface frontier has no obvious change.

1. Introduction
Phase field method\textsuperscript{[2-4]} is one of the widely applied computer numerical simulation techniques. Based on Ginsberg-Landau theory\textsuperscript{[5]}, it coupled the combined effects of diffusion, ordering potential, thermodynamics and dynamics through differential equations. The solution of the phase field method can describe the liquid-solid interface morphology and interface migration in metals and alloy systems, thereby avoiding tracking the complex solid-liquid interfaces and directly simulating microstructures formation. It has unique advantages in the numerical simulation of metal solidification processes.

The relevant scholars in the world have adopted the phase field method to conduct extensive research on the eutectoid transformation\textsuperscript{[6-7]}. Feng Li\textsuperscript{[8]} used the phase field method to simulate the growth morphology of the lamellar pearlite microstructures with different initial undercooling and the different initial lamella spacing; Kumar\textsuperscript{[9]} used analytical methods to study the effect of diffusion of growth phase on the growth rate of the lamellar front edge, and compared the analytical prediction results with the phase field numerical simulation results, which was well confirmed; Katsumi\textsuperscript{[10]} by considering the diffusion of carbon in both austenite and ferrite, use phase-field method to studied the cooperation growth of ferrite and cementite during the pearlite transformation. At the same time, predicted the lamellar spacing and growth rate of the lamellar pearlite microstructure under different undercooling, and the results were compared with the experimental results; Steinbach\textsuperscript{[11]} studied the effect of stress and strain on pearlite transformation kinetics by the phase field method. In summary, related scholars have less research in the influence of interfacial energy and diffusion coefficient on the lamellar pearlite microstructure. However, the formation of pearlite lamellar is greatly affected by the interfacial energy and the diffusion coefficient.

In this paper, pearlite eutectoid steel is taken as an example. The KKSO phase-field model coupled
concentration field equation is used to simulation study the evolution of the microstructure and the
distribution of solute of lamellar pearlite in different diffusion coefficients and different interfacial
energies.

2. Phase-field model

2.1. Phase-field equation

The phase field control equation adopts the KKSO model [1], and its phase-field control equation is:

$$\frac{\partial \phi_i}{\partial t} = -\frac{2}{n} \sum_j S_{ij} M_{ij} \left[ \frac{\partial F}{\partial \phi_i} - \frac{\partial F}{\partial \phi_j} \right]$$  \hspace{1cm} (1)

In Eq. 1,

$$\frac{\partial F}{\partial \phi_i} = \sum_{j \neq i} \left[ \frac{\varepsilon_{ij}^2}{2} \nabla^2 \phi_j + w_{ij} \phi_j \right] + f^i(c_i) - c_i f_c$$  \hspace{1cm} (2)

In Eq. 2,

$$f^i(c_i) - c_i f_c = -\frac{RT_E}{V_m} \left( c_E - \frac{T_e - T}{m_i} \right)(1 - k_i) - \frac{RT_E}{V_m} c_i \quad (i = 1, 2)$$  \hspace{1cm} (3)

When \( i = 3 \),

$$f^3(c_3) - c_3 f_c = -\frac{RT_E}{V_m} c_3$$  \hspace{1cm} (4)

In Eq. 1 and Eq. 2: phase field order parameter \( \Phi_i \) (\( i = 1, 2, 3 \)), which represents the volume fraction of ferrite, cementite, and austenite at any position during solidification. The three order parameters at any position in the solidification process follow the following rules:

$$\sum_i \phi_i = 1 \quad (i = 1, 2, 3) \quad n = \sum_i s_i \quad s_{ij} = s_i s_j \quad S_i \quad M_{ij}$$

\( M_{ij} \) is a phase field dynamic coefficient and \( M_{ij} = \frac{V_m}{RT} \left[ \frac{\sigma_{ij}}{2} \right] \left[ \frac{1 - k_i}{c_{ij}^e \sigma_{ij}^e} \right] \). The superscript \( e \) represents the equilibrium state; \( \varepsilon_{ij} \) is the gradient energy coefficient and its expression is \( \varepsilon_{ij} = \frac{4}{\pi} \sqrt{\sigma_{ij}} \); \( w_{ij} \) is the interface thickness parameter and \( w_{ij} = 2 \sigma_{ij} / \zeta \); \( \zeta \) is half of the interface thickness; \( \sigma \) is the interface energy; \( f \) is the free energy density; \( c \) is the component of the system. In Eq.3: \( T_e \) is the eutectoid temperature; \( C_E \) is the eutectoid component; \( k_i \) is the equilibrium distribution coefficient; \( m_i \) is the slope of the equilibrium parent phase.

2.2. Solute field equation [8]

$$\frac{\partial C}{\partial t} = \nabla \cdot D \sum_i \phi_i \nabla c_i$$  \hspace{1cm} (5)

Where, \( D \) is the solute diffusion coefficient; \( \Phi_i \) is the phase field order parameter; \( c_i \) (\( i = 1, 2, 3 \)) represents the solute concentration of ferrite, cementite and austenite in the solidification process.
3. Numerical calculation of the model

3.1. Initial conditions and boundary conditions
In this paper, Fe-3.494 %C binary alloy is used as an example to study and the equilibrium transition temperature is 1000K. Under isothermal conditions (initial undercooling is 20K), simulation the eutectoid transformation process. In the simulation study, the minimum undercooling lamellar spacing [8] was used as the calculation benchmark, the initial pearlite lamellar is introduced at the bottom of the calculation area (In Fe-C alloy phase diagrams, the lamellar width ratio of ferrite and cementite is calculated using the law of leverage to be 7:1 approximately), The left and right boundaries are set as periodic boundary conditions, and the upper and lower boundaries are set as adiabatic boundary conditions.

3.2. Numerical calculation method
The explicit finite difference method is used to solve Eq.1 and Eq.5 at the same time step $\Delta t$. The time step $\Delta t$ and the space step size $\Delta x$ need to satisfy the following stable conditions:

$$
\Delta t \leq \left(\frac{\Delta x}{4D}\right)^2
$$

(6)

3.3. Physical parameters of the material

Table 1. Material physical parameters of Fe-C binary alloy [12-13].

| Parameter                          | Value          |
|-----------------------------------|----------------|
| Ferrite distribution coefficient $k_1$ | 0.9228         |
| Cementite distribution coefficient $k_2$ | 1.1081         |
| Ferrite diffusion coefficient $D_f$ [m$^2$/s] | 2.2435x10$^{-11}$ |
| Austenite diffusion coefficient $D_a$ [m$^2$/s] | 1.0145x10$^{-12}$ |
| Ferrite phase slope $m_1$ [K/mol%] | -52.9378       |
| Cementite phase slope $m_2$ [K/mol%] | 74.1336       |
| Ferrite/cementite interface energy $\sigma_{12}^0$ [J/m$^2$] | 0.9223         |
| Ferrite/austenite interface energy $\sigma_{13}^0$ [J/m$^2$] | 0.85           |
| Cementite/austenite interface energy $\sigma_{23}^0$ [J/m$^2$] | 0.68           |
| Molar volume $V_m$ [m$^3$/mol] | 7.7x10$^{-6}$  |

4. Results and analysis

4.1. Morphology and Solute distribution of eutectoid lamella under different interfacial energy conditions
Figure 1 show the growth morphology of lamellar pearlite microstructure at different interfacial energies. Figure 1 (a-e) represents the pearlite growth morphology with interfacial energy of $\sigma_y = \sigma_y^0$, $\sigma_{ij} = 1.3\sigma_y^0$, $\sigma_{ij} = 1.5\sigma_y^0$, $\sigma_{ij} = 1.7\sigma_y^0$, $\sigma_{ij} = 1.9\sigma_y^0$ ($\sigma_y$ include $\sigma_{12}$, $\sigma_{13}$, $\sigma_{23}$), respectively, when the initial lamellar spacing is the equilibrium lamellar spacing $\lambda_0 = 0.4m$ [8] and the initial undercooling is 20K. In Figure 1, the value of the phase field order parameter is shown as a scale.
Figure 1. Growth morphology of lamellar pearlite microstructure at different interface energies.

Figure 1 (a-e) is the growth morphology of lamellar pearlite microstructure under the same time step and different interfacial energy. It can be seen that with the increase of interfacial energy, the lamellar growth pattern of pearlite change from the regular symmetry to low symmetry and small amplitude oscillation. At the same time, the growth rate of pearlite decreases with the increase of interface energy. Observing the interface frontier morphology of the pearlite under different interface energy conditions, it can be seen that the morphology of the front edge of the ferrite phase interface changed convex from the horizontal with the interface energy increases. According to the Gibbs free energy principle, as the interfacial energy increases, the pressure on both sides of the interface frontier becomes larger, so that the center of curvature of the interface gradually transitions from the austenite phase to the pearlite, and the interface appears above process of the change from horizontal to the convex.

4.2. Morphology and solute distribution of eutectoid lamella under different diffusion coefficient conditions

Figure 2 and Figure 3 respectively show the growth morphology and solute distribution of lamellar pearlite. Figure 2 (a-e) represents the growth morphology of lamellar pearlite when the diffusion coefficients of carbon atoms in austenite are $0.3 \times 10^{-12} \text{m}^2 \cdot \text{s}^{-1}$, $0.7 \times 10^{-12} \text{m}^2 \cdot \text{s}^{-1}$, $1.0145 \times 10^{-12} \text{m}^2 \cdot \text{s}^{-1}$, $1.2 \times 10^{-12} \text{m}^2 \cdot \text{s}^{-1}$, $1.5 \times 10^{-12} \text{m}^2 \cdot \text{s}^{-1}$, respectively (the diffusion coefficient changes with temperature). Figure 3(a-e) is a diagram of the solute distribution corresponding to the growth of pearlite in Figure 2 (a-e). In Figure 2, the value of the phase field order parameter is shown as a scale; in Figure 3, the solute content value is shown as a scale.

Figure 2. Growth morphology of lamellar pearlite microstructure at different diffusion coefficients.
Figure 3. The solute distribution of lamellar pearlite microstructure at different diffusion coefficients.

Figure 2 (a-e) shows the growth morphology of pearlite at the same time step. It can be seen that the growth rate of pearlite gradually increases with the increase of diffusion coefficient. Observing the frontier of the pearlite growth interface, it was found that the frontier morphology of the pearlite interface did not obvious change with the increase of the diffusion coefficient. This is because $D_a \ll D_f$, changing $D_a$ does not cause the interface to destabilize, and therefore the morphology of the interface frontier will not have much impact. Figure 3 (a-e) shows the solute distribution of pearlite microstructure with different diffusion coefficients. The solute distribution in the Figure 3 is consistent with the growth morphology of its eutectoid lamellar. As the diffusion coefficient increases, the solute diffusion capacity increases, and the components required for the growth of ferrite and cementite lamellar are rapidly and effectively replenished. Therefore, the growth rate of lamellar pearlite tends to increase.

5. Conclusions
(1) With the interfacial free energy increases, the lamellar pearlite growth mode changes from regular symmetry to low-symmetry, small-amplitude lamellar growth mode, the pearlite growth rate decreases and the front morphology of the ferrite phase interface changes from horizontal to convex.

(2) With the diffusion coefficient increases, the growth rate of lamellar pearlite gradually increases and the frontal morphology of the lamellar pearlite interface does not obvious change.

6. Acknowledgement
This work is supported by The National Natural Science Foundation of China (Grant nos: 11504149, 11364024, and 51661020).

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