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Journal of Crystal Growth 198/199 (1999) 1275–1280

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JOURNAL OF **CRYSTAL  
GROWTH**

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# Phase field simulations of the peritectic solidification of Fe–C

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## Abstract

The microstructure evolution during the peritectic solidification of Fe–C is simulated using a multiphase field approach. The peritectic transformation in steel is a diffusion controlled non-equilibrium process with the three phases liquid, ferrite and austenite involved. Phase fields are defined for each of the three phases. A set of phase field equations is derived from a free energy formulation, whereas the carbon diffusion equation is derived separately by a solute diffusion model. Both phase field and solute diffusion model as well as their coupling will be discussed. Simulation results will be presented showing the growth of the primary ferrite and the subsequent peritectic transformation, in which the austenitic phase is formed. © 1999 Elsevier Science B.V. All rights reserved.

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## 1. Introduction

Steels with low carbon concentration solidify in a peritectic transformation [1,2]. The primary solid phase which forms from the melt is the  $\delta$ -ferrite. This phase has a low carbon solubility and usually solidifies in dendritic morphology. Below the peritectic temperature ( $\approx 1766$  K) the ferrite reacts with the residual melt to form the  $\gamma$ -austenite. Assuming moderate cooling rates, both the growth of the ferrite phase and the subsequent peritectic transition are diffusion controlled processes. Thermodynamic equilibrium conditions can only be assumed at the phase interfaces. The binary phase diagram of Fe–C with its metastable extensions is shown in Fig. 1. In order to predict the microstructure evolution during the described transformations, a model has to consider the following aspects:

- multiphase systems
- non-equilibrium and metastable equilibrium conditions
- dendritic growth
- nucleation of new phases
- solute diffusion
- coupling between phase transformations and solute diffusion.

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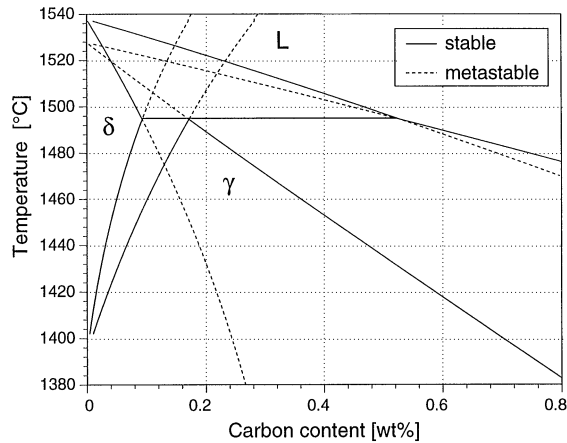


Fig. 1. Fe–C phase diagram with metastable extensions [8].

A microstructure simulation model based on the phase field method which accounts for these aspects in binary alloys has recently been developed by ACCESS [3]. The principles of this model will be discussed in the following section. Simulation results of the peritectic solidification of Fe–C are presented in Section 3.

## 2. The multi phase field approach with an integrated model for solute diffusion

In the standard formulation of the phase field method, the structure of a system is described by a single phase field  $\phi(\mathbf{x}, t)$ , which is defined as a continuous function in space and time.  $\phi = 0$  denotes “phase A” (e.g. liquid) and  $\phi = 1$  denotes “phase B” (e.g. solid) [4,5]. In order to extend the phase field method to polyphase solidification, a multiphase field model has been developed [6]. This model is based on multiple phase fields  $\phi_i(\mathbf{x}, t)$ ,  $i = 1, \dots, n$  for  $n$  different phases, e.g. in the case of peritectic solidification of steel for the liquid, the ferrite and the austenite. In polycrystal simulations also different grains of the same phase may be distinguished by individual phase fields. In the multiphase model  $\phi_i = 1$  denotes “phase  $i$ ” and  $\phi_i = 0$  “not phase  $i$ ”. As a natural constraint, the sum of the phase fields equals 1. In the diffuse interface regions, the phase fields vary continuously between 0 and 1 as known from the standard phase field model.

A local free energy functional  $\hat{f}$  is defined with  $\sigma_{ik}$  being the interfacial energies,  $\eta_{ik}$  the interfacial thicknesses,  $\Delta S_{ik}$  the entropy of fusion and  $\Delta T_{ik}$  the local undercooling with respect to the phases  $i$  and  $k$ .

$$\hat{f} = \sum_{\substack{i < k \\ i, k}}^n \left( \frac{\varepsilon_{ik}^2}{2} \left| \phi_k \nabla \phi_i - \phi_i \nabla \phi_k \right|^2 + \frac{1}{4a_{ik}} [\phi_i^2 \phi_k^2 - m_{ik} (\frac{1}{3} \phi_i^3 + \phi_i^2 \phi_k - \frac{1}{3} \phi_k^3 - \phi_k^2 \phi_i)] \right), \quad (1)$$

$$\varepsilon_{ik}^2 = \eta_{ik} \sigma_{ik}; \quad a_{ik} = \frac{\eta_{ik}}{72 \sigma_{ik}}$$

and

$$m_{ik} = 6a_{ik} \Delta S_{ik} \Delta T_{ik}(c, T).$$

The gradient term in the free energy formulation drives the interface to a diffusive state. This term is also sensitive on curvature. The gradients are weighted by the density of the counter phases in order to distinguish the contributions of the different dual phase combinations  $ik$ . Each contribution  $ik$  is related to an individual  $\varepsilon_{ik}$  and equals zero if one of the phases is absent. The second term determines the potential energy including the thermodynamic driving force  $m_{ik}$ .

Minimization of the functional (1) and definition of kinetic parameters  $\tau_{ik} = \Delta S_{ik} \eta_{ik} / \mu_{ik}$  with  $\mu_{ik}$  being the interface mobility leads to the kinetic equations:

$$\phi_i = \sum_{\substack{k \\ k \neq i}}^n \frac{1}{\tau_{ik}} \left[ \varepsilon_{ik}^2 (\phi_k \nabla^2 \phi_i - \phi_i \nabla^2 \phi_k) - \frac{\phi_i \phi_k}{2a_{ik}} (\phi_k - \phi_i - 2m_{ik}) \right]. \tag{2}$$

A major advantage of this approach is, that it allows the specification of the physical parameters ( $\sigma_{ik}, \Delta S_{ik}, \Delta T_{ik}, \mu_{ik}$ ) individually for each interface. In this formulation triple points are described by superposition of pairwise interactions of the phases. A rigorous treatment of multiphase systems would lead to additional triple point contributions, which are neglected here. This is discussed in Ref. [7]. As known from other phase field models, anisotropy can be considered by orientation dependent kinetic parameters or by orientation dependent interfacial energies. Seeds of new phases are generated as function of a critical local undercooling and a random term by an additional nucleation model.

Solute diffusion is calculated within the whole multiphase system. To allow compatibility with the multi-phase-field approach, the diffuse interfaces regions where two or more phases coexist are taken into account by defining a mixture concentration:

$$c(\mathbf{x}, t) = \sum_i^N \phi_i(\mathbf{x}, t) \cdot c_i(\mathbf{x}, t) \tag{3}$$

with  $c_i$  being the concentration of a component in phase  $i$ . In the bulk of phase  $i$ , the concentrations  $c$  and  $c_i$  are identical, whereas at the interface with a second phase  $j$  the mixture concentration  $c$  changes steeply but continuously from  $c_i$  to  $c_j$ . Within the diffuse interface areas the concentrations are related by the equilibrium partition coefficients  $k_{ij}$ , which usually are a function of temperature:

$$c_i(\mathbf{x}, t) = k_{ij}(T) \cdot c_j(\mathbf{x}, t). \tag{4}$$

The partition coefficients are defined by the partition of the equilibrium concentrations. For the non-equilibrium state we locally assume the same partition for all concentrations within the interface. This corresponds in first order approximation with the same degree of local undercooling for both phases  $i$  and  $j$ ;  $\Delta_T(c_i) = \Delta_T(c_j)$ . If the interface thickness is small compared to the diffusion length, Eq. (4) corresponds with the assumption of local equilibrium at the interface in a sharp interface model.

To describe solute transport within different phases  $i, i = 1, \dots, n$  with changing solubilities and diffusion coefficients  $D_i$  as well as across the phase boundaries we take the following approach:

$$\dot{c}(\mathbf{x}, t) = \nabla \left( \sum_{i=1}^n \phi_i D_i \nabla c_i \right). \tag{5}$$

Choosing arbitrarily a reference phase  $R$ , obeying  $\phi_R > 0$ , all  $c_i$  can be replaced by the partition coefficients with respect to this reference phase  $k_{iR}$  multiplied by the concentration of the reference phase:

$$\begin{aligned} \dot{c}(\mathbf{x}, t) &= \nabla \left( \sum_{i=1}^n \phi_i D_i \nabla (k_{iR} c_R) \right) \\ &= \nabla \left( \sum_{i=1}^n \phi_i D_i \nabla \frac{k_{iR} c}{\sum_j \phi_j k_{jR}} \right). \end{aligned} \tag{6}$$

It shall be emphasized that in this formulation solute redistribution due to continuous interface movement is treated implicitly. No additional flux balances or source terms are required at the interfaces. The phase field equations (Eq. (2)) and the concentration equation (Eq. (6)) are coupled by the common phase field parameters  $\phi_i$ , and the concentration dependent local undercooling term  $\Delta T(c, T)$ . The undercooling is defined by the deviation from the local equilibrium temperature  $T_{ij}^*$ , which can be deduced from the specific phase diagram. In linear approximation we get

$$T_{ij}^* = T_{ij}^{\circ} + m_{ij}c_i = T_{ij}^{\circ} + m_{ij} \frac{c}{\sum_l \phi_l k_{lR}}. \quad (7)$$

Since the driving force should essentially be constant across the diffuse interface, high concentration gradients may lead to numerical problems. For this reason the interface thickness has to be small compared to the solutal diffusion length. However, this assumption often requires a very fine discretisation requiring high computation times. To diminish this problem we average the driving force across the interface in direction of the phase field gradient.

### 3. Simulations of the peritectic solidification of FeC

2D-simulations of the peritectic solidification of FeC have been carried out using the model described in Section 2. The parameters for the calculation shown in Fig. 2 are  $c_0 = 0.3$  wt%,  $\dot{T} = 0.5$  K/s,  $D_L = 3 \times 10^{-5}$  cm<sup>2</sup>/s,  $D_\delta = 6 \times 10^{-6}$  cm<sup>2</sup>/s,  $D_\gamma = 10^{-6}$  cm<sup>2</sup>/s,  $\sigma_{ij} = 2.04 \times 10^{-4}$  J/cm<sup>2</sup>. Thermodynamic data are taken in linear approximation from the metastable phase diagram of Fe–C depicted in Fig. 1. A grid of  $150 \times 150$  cells with a spacing of 2  $\mu$ m and a time step of  $\nabla t = 5 \times 10^{-4}$  s are used and periodical boundary conditions are applied. The growth of four ferritic particles during constant cooling has been simulated and below the peritectic temperature single nuclei have been placed onto the interfaces to study the peritectic transformation. The distances of the particles have been selected in the order of typical secondary dendrite arm spacings and anisotropy has been not considered in this calculation. During the growth of the ferritic

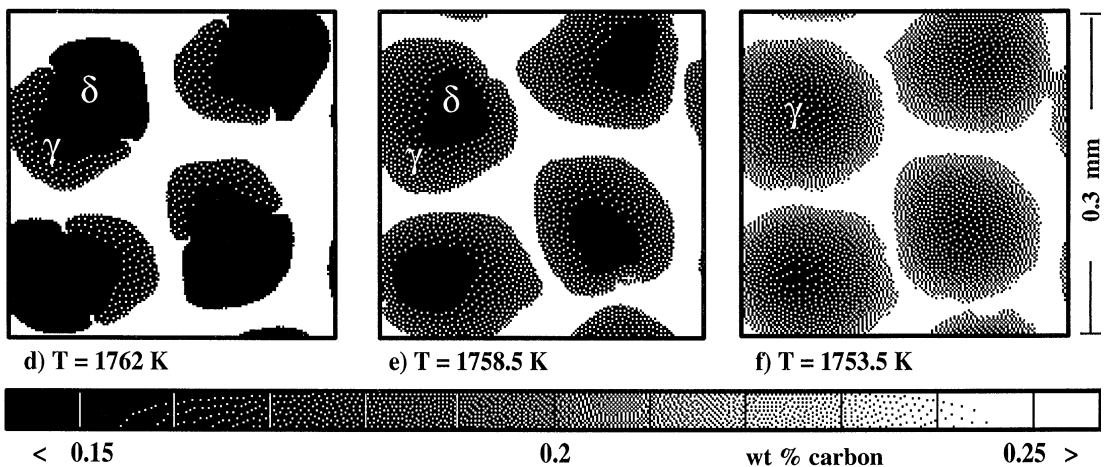


Fig. 2. Simulation of the peritectic solidification in Fe–C. The carbon concentration is illustrated by the grey scale. The peritectic  $\gamma$ -austenite nucleated on the primary  $\delta$ -ferrite grows around it by simultaneous consumption of both the ferrite and the liquid.

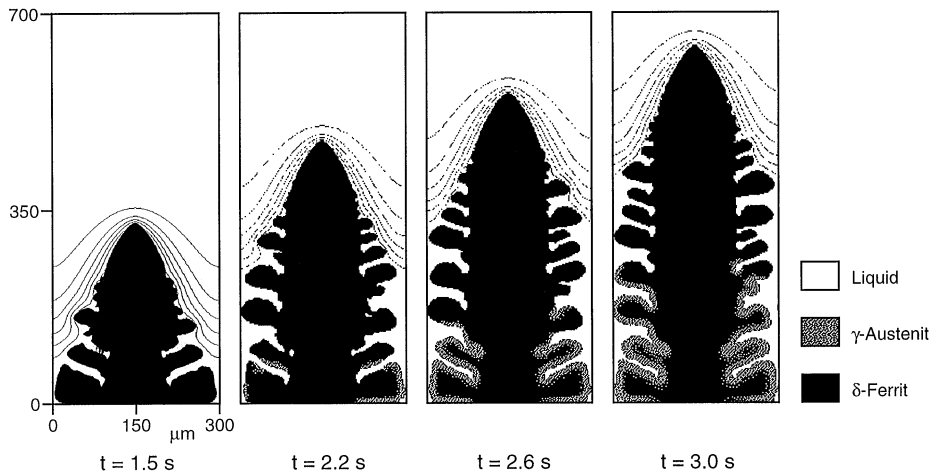


Fig. 3. Simulation of a directional peritectic solidification process. The morphology of the primary ferrite becomes unstable and a dendrite evolves. Beyond the peritectic temperature austenite is formed and coats the dendrite.

particles the melt becomes enriched with carbon due to the low carbon solubility of the ferrite phase. Since the carbon content of the austenite ( $\approx 0.16$  wt%) at this temperature is higher than the ferrite's concentration ( $\approx 0.1$  wt%) but less than the liquid's concentration ( $\approx 0.51$  wt%), the austenite grows fastest where liquid and ferrite can react in direct contact. Thus, the austenite grows around the ferrite. This is the typical peritectic behaviour known from both experiment and theory [1,2]. When the austenite isolates the ferrite from the liquid, ferrite and liquid can no longer react in this direct kind of peritectic reaction. However, the peritectic transition continues indirectly by solid state diffusion through the austenite layer.

In another simulation (Fig. 3) directional growth is investigated with an imposed temperature gradient of  $\nabla T = 140$  K/cm and a cooling rate  $\dot{T} = -3.0$  K/s on a grid with  $300 \times 700$  cells. Anisotropy is now considered by an orientation dependent kinetic parameter  $\tau_{ik}(\theta)$ . During the growth of the ferrite the redistribution of carbon leads to morphological instability and a dendrite evolves. Below the peritectic temperature the austenitic seeds are generated by a nucleation model in dependence on the local undercooling and a random term. The austenite grows around the undercooled part of the ferritic dendrite, consuming both the liquid and the ferrite.

#### 4. Conclusions

A multiphase phase field method with an integrated diffusion model for binary alloys has been developed and applied to simulate the microstructure evolution during the peritectic transformation of Fe–C. The model enables the specification of individual physical parameters for all three phases (liquid, ferrite, austenite) and their interface combinations (liquid/ferrite, liquid/austenite, ferrite/austenite). In order to simplify considerations of complex phase diagrams, the solute diffusion equation is not derived from the free energy functional but described using partition coefficients at the interfaces. The driving force for phase transitions is defined by the local undercooling, which is dependent on the local concentrations within the different phases.

Simulation of the peritectic solidification of FeC have been carried out for realistic cooling conditions. The simulation results reproduce well the expected peritectic phenomena and illustrate the strong interaction between carbon redistribution and peritectic phase transformation. Dendritic growth of the primary ferrite

phase has been simulated choosing physical instable conditions. However, high concentration gradients within the diffuse interface regions still lead to numerical problems. In order to avoid these instabilities the interface thickness has to be small compared to the solutal diffusion length. Therefore, dendrites with low supersaturations require very high numerical resolution leading to high CPU-times. A first step to overcome this problem was the definition of an average driving force across the interface in direction of the phase field gradient.

In related activities the same model has successfully been applied to simulate different aspects of eutectic solidification [9,10].

### **Acknowledgements**

The work was supported by the German Bundesministerium für Bildung, Wissenschaft, Forschung und Technologie under grant number 03 IX 80027 (COST 512).

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