

# Coupled and competitive eutectic growth

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## Abstract

A description of the competitive growth is presented in the paper. The description is associated with the competition between eutectic structure and primary phase formation. A coupled zone for the eutectic solidification is drawn in the phase diagrams. The coupled zone is shown as a range of solute concentration versus under-cooling to justify the formation of the eutectic structure, exclusively. Interface growth temperatures of the single and coupled eutectic are illustrated schematically as a function of growth rates at a given composition  $C_0$ .

**Keywords:** Eutectic solidification, Kinetics, Competitive growth, Coupled growth

## 1. Introduction

Evolution of solidification microstructures can be the strategic link between materials processing and materials behavior. The eutectic structure is the basis of most commercial casting alloys, and thus, the properties of these alloys strongly depend on the amount and morphology of the eutectic phases, which, in turn, are affected by various variables, including cooling rate, modification, and faceted or nonfaceted nature of the constituent phases [1].

Basic concepts of the theory of eutectic alloys are a small but theoretically and practically important part of the science of metals. In accordance with the old and universally accepted ideas the components of eutectic systems, which are almost insoluble (in many systems) in solid state, are infinitely miscible in the liquid state, i.e., at a temperature above the liquidus line on the phase diagram alloys are treated as liquid solutions of components. When cooled to the eutectic temperature  $T_e$ , a solution becomes supersaturated with both components; its crystallization occurs by diffusion decomposition into a mixture of crystals of almost pure components (solid solutions on their base, i.e.,  $\alpha$  and  $\beta$ ). Eutectic equilibrium is described as  $L = \alpha + \beta$ .

Directional solidification of binary or pseudo-binary eutectics, may result in regular structures of fibrous or lamellar type. Typi-

cal eutectic structures of binary alloys form by the simultaneous growth of two phases from the liquid; therefore they may exhibit a variety of microstructures that can be classified according to two criteria:

- lamellar vs. fibrous morphology of the individual phases, and
- regular vs. irregular growth of the individual phases.

Directional solidification technique is widely applied not only to the research on the basic solidification theory, but also to manufacturing high-performance products, such as superalloy turbine blades with single crystal structure in aeronautical engines.

## 2. Competitive growth in eutectic alloys

A competitive growth mechanism of eutectic has been suggested by Tammann and Botschwar [2] from a study of formation ability of eutectic structures. That is to say, the microstructure of alloys near the eutectic point is determined by competition between the eutectic structure and the primary phase. Only when the growth of phases takes precedence over that the primary phase, the eutectic-like microstructure can be produced. Otherwise, the primary phase is dominant.

In directional solidification, the interface growth temperature of the primary phase at different growth rates can be described by [3]:

$$T_j^i = T_L^i - \frac{G_L D_L}{V} - A_j V^{1/2}, j = \alpha, \beta \quad (1)$$

where:

$T_L^i$  - the liquidus temperature at the alloy composition  $C_o$ ,  
 $V$  - the growth rate,  
 $G_L$  and  $D_L$  - the temperature gradient and diffusion coefficient in liquid, respectively.

The parameter  $A_j$  in eq. (1) is given as:

$$A_j = \left[ -\frac{8\Gamma_j m_j C_o (1-k_j)}{D_L} \right]^{1/2}, j = \alpha, \beta \quad (2)$$

where:

$\Gamma_j$ ,  $m_j$  and  $k_j$  - Gibbs-Thomson coefficient, slope of  $j$ -phase liquidus and solute distribution coefficient, respectively.

For eutectic solidification, the interface growth temperature of coupled eutectic in directional solidification can be calculated using the model described by Magnin and Trivedi [4] as:

$$T_e^i = T_E - (\phi + 1/\phi)(K_1 K_2)^{1/2} V^{1/2} = T_E - B V^{1/2}, \quad (3)$$

$$\text{where } B = (\phi + 1/\phi)(K_1 K_2)^{1/2}, K_1 = \frac{\bar{m} C_e^o P}{D_L f_\alpha f_\beta}, \quad (4)$$

$$K_2 = 2\bar{m}\delta \left( \frac{\Gamma_\alpha \sin\theta_\alpha}{|m_\alpha| + m_\beta} + \frac{\Gamma_\beta \sin\theta_\beta}{m_\beta f_\beta} \right), \quad (5)$$

$$\bar{m} = \frac{|m_\alpha| m_\beta}{|m_\alpha| + m_\beta}, \quad (6)$$

$$C_e^o = C_\beta^o - C_\alpha^o. \quad (7)$$

For lamellar eutectic growth, the parameters  $P$  and  $\delta$  can be written in simplified forms as:

$$P \cong 0,3383 (f_\alpha f_\beta)^{1,661}, \quad (8)$$

$$\delta = 1. \quad (9)$$

For rod eutectic growth, the parameters  $P$  and  $\delta$  can also be simplified as:

$$P \cong 0,167 (f_\alpha f_\beta)^{1,25}, \quad (10)$$

$$\delta = 2\sqrt{f_\alpha}, \quad (11)$$

where:

$T_E$  - the equilibrium solidification temperature of eutectic,

$\bar{m}$  - the average slope of eutectic defined in eq. (4),

$f_\alpha, f_\beta$  - the volume fractions of  $\alpha$  phase and  $\beta$  phase respectively,

$C_e^o$  - the composition difference between the solubility limits of  $\beta$  phase ( $C_\beta^o$ ) and  $\alpha$  phase ( $C_\alpha^o$ ) defined in eq. (7) at the eutectic solidification temperature [4].

A methodology of competitive growth outlined above provides an adequate framework to understand the major features of the transition from eutectic to dendritic growth. However more subtle variations in eutectic microstructure occur under conditions close to the transition that require a more complete analysis of interface stability [6].

### 3. Prediction of eutectic coupled zone

The argument in this respect is whether the thermal undercooling  $\Delta T_t$  and kinetic undercooling  $\Delta T_k$  can be omitted when dealing with the lamellar eutectic growth within an undercooled alloy melt. The bulk undercooling  $\Delta T$  of a liquid alloy is usually divided into four parts [7,9]:

$$\Delta T = \Delta T_c + \Delta T_r + \Delta T_t + \Delta T_k \quad (12)$$

where:

$\Delta T_c$  and  $\Delta T_r$  - the solute undercooling and curvature undercooling,  $\Delta T_t$  - thermal undercooling,  $\Delta T_k$  - kinetic undercooling.

So far, there has been no analytical model to specify the thermal undercooling for eutectic growth. If the bulk undercooling is not very large,  $\Delta T_c$  and  $\Delta T_r$  play the dominant roles, whereas  $\Delta T_t$  and  $\Delta T_k$  only make minor contributions.

This has been confirmed by the experimental work of many investigators. In fact, there exist two undercooling thresholds for the "lamellar eutectic-anomalous eutectic" structural transition [10]. Below the lower undercooling threshold  $\Delta T_1^*$  of about 30–60 K, lamellar eutectic is the unique growth morphology. Above the upper undercooling threshold  $\Delta T_2^*$  of about 150–200 K, only anomalous eutectic can grow. In the intermediate undercooling regime of  $\Delta T_1^* - \Delta T_2^*$ , both lamellar eutectic and anomalous eutectic coexist. Metallographic analyses demonstrate that anomalous eutectic is the product of rapid solidification during recalescence, while lamellar eutectic forms in the slow period of solidification after recalescence. Because the remnant undercooling at the end of recalescence becomes quite small, lamellar eutectic growth corresponds to the small undercooling condition even within the intermediate undercooling regime.

As a first order approximation, it is reasonable to neglect the influences of thermal undercooling  $\Delta T_t$  and kinetic undercooling  $\Delta T_k$  on lamellar eutectic growth. Consequently, eq. (1) leads to the following approximate relation [7,8,9]:

$$\Delta T \approx \Delta T_c + \Delta T_r \quad (13)$$

### 4. Coupled growth

The coupled growth zone marks the range of the chemical composition, the growth rate and the temperature gradient,

which assure the obtainment of the exclusively eutectic structure (without hypoeutectic phase). Way of marking coupled zone on the basis of the theory of the competitive growth was showed on the figure 2 [9, 10].

Figure 3 is a kind of phase diagram in eutectic systems that the coupled zone encompasses the  $\alpha$  liquidus extension. Interface growth temperatures of the single  $\alpha$  phase, single  $\beta$  phase and coupled eutectic ( $\alpha+\beta$ ) calculated by eq. (1) and eq. (3) are illustrated schematically as a function of growth rates at a given composition  $C_0$  in hypereutectic alloy on the right side of figure 3.

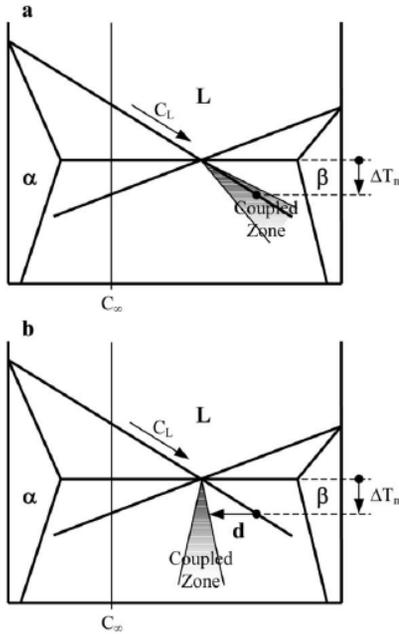


Fig. 2. (a) The coupled zone encompasses the  $\alpha$  liquidus extension: coupled eutectic growth occurs directly from the primary  $\alpha$  dendrites. (b) The coupled zone does not encompass the  $\alpha$  liquidus extension: haloes of  $\beta$  form around the primary  $\alpha$  dendrites for any significant  $\beta$  nucleation undercooling  $\Delta T_n$  [8,9,11]

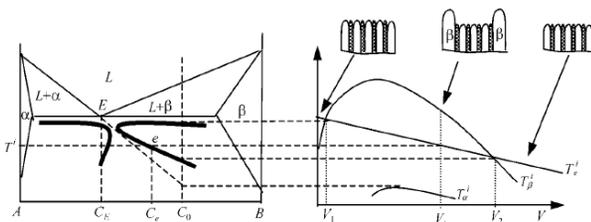


Fig. 3. Coupled zone encompasses the  $\alpha$  liquidus extension in eutectic systems and interface growth temperatures of the single  $\alpha$  phase, single  $\beta$  phase and coupled eutectic ( $\alpha+\beta$ ) are calculated by eqs. (1) (3) as a function of growth rates at a given composition  $C_0$  in a hypereutectic alloy [5]

Coupled growth zone can be outlined with the same interface growth temperature and composition for the single phase and coupled eutectic. From figure 3, at lower growth rate  $V_1$  and

higher growth rate  $V_2$ , the interface temperature of the single  $\beta$  phase  $T_\beta^i$  and coupled eutectic  $T_e^i$ , are equal at the given composition  $C_0$ . When the imposed growth rate  $V_i$  is below  $V_1$ , coupled eutectic ( $\alpha+\beta$ ) becomes stable due to the sharp drop in the single-phase temperature, which contributes to the presence of the positive gradient through the term,  $G_L D_L / V$  in eq. (1). Thus for finite  $G_L$ , the single-phase interface temperature is given by eq. (1) in which the contribution from the third term on the right hand side is negligible. Eq. (1) and eq. (3) can be simplified at low growth rate  $V_1$  as:

$$T_\beta^i \approx T_L^\beta - \frac{G_L D_L}{V} - A_\beta V^{1/2} \approx T_E + m_\beta (C_0 - C_E) - \frac{G_L D_L}{V}, \quad (14)$$

$$T_e^i = T_E - B V^{1/2} \approx T_E, \quad (15)$$

where:

$C_E$  - the eutectic composition, by equating eqs. (14) and (15), the value of the growth rate  $V_1$ , at which the primary  $\beta$  phase-coupled eutectic ( $\alpha+\beta$ ) transition occurs at low undercoolings can be derived as:

$$V_1 = \frac{G_L D_L}{m_\beta (C_0 - C_E)}. \quad (16)$$

At high growth rate, the term  $G_L D_L / V$  in eq. (1) is small and can be neglected. Under this assumption, eq. (14) can be approximated as:

$$T_\beta^i \approx T_L^\beta - \frac{G_L D_L}{V} - A_\beta V^{1/2} \approx T_E + m_\beta (C_0 - C_E) - A_\beta V^{1/2}. \quad (17)$$

Equating eq. (3) and eq. (17), the value of the high growth rate  $V_2$  at which the primary  $\beta$  phase coupled eutectic ( $\alpha+\beta$ ) transition occurs at high undercoolings shown in figure 3 can be written as:

$$V_2^{1/2} = \frac{m_\beta (C_0 - C_E)}{A_\beta - B} \quad (18)$$

The right hand side of eq. (18) should be positive and values of  $A_\beta$  and  $B$  can be calculated using eq. (2) and eq. (3), respectively.

From eq. (16) and eq. (18), with the composition  $C_0$  approaching the eutectic composition  $C_E$ , the value of the low growth rate  $V_1$  increases and the value of the high growth rate  $V_2$  decreases. If  $V_1$  is equal to  $V_2$ , the alloy composition  $C_0$  corresponding to coupled eutectic ( $\alpha+\beta$ ) growth at any growth rates can be obtained in directional solidification. Moreover, with the increasing growth rate, coupled eutectic ( $\alpha+\beta$ ) will be refined as shown in figure 3.

In addition, if the imposed growth rate is very large, it should be noted that the directional heat flux would be destroyed and the solidified microstructure would not be the directionally coupled eutectic and it may be the equiaxed eutectic [5].

Figure 4 shows that the  $\alpha$  liquidus extension is not encompassed in the coupled zone. At the growth rate  $V_{c1}$ , shown in the right side of figure 4, the composition of the  $\alpha$  liquidus is  $C_b$  (here  $C_b$  is equal to  $C_0$ ) at the nominal alloy composition  $C_0$ . With the imposed growth rate increasing from  $V_{c1}$  to  $V_{c2}$ , the composition of coupled eutectic ( $\alpha+\beta$ ) changes from  $C_e$  to  $C_{e'}$  and simultane-

ously, the composition of  $\alpha$  liquidus changing from  $C_b$  to  $C_{b'}$  shown in the left part of figure 4. Thus at the growth rate  $V_{c2}$ , the single  $\alpha$  phase corresponding to the nominal alloy composition  $C_0$  will lead the coupled eutectic ( $\alpha+\beta$ ) growth with some undercooling  $\Delta T_n$ , seen in figure 2. If the undercooling  $\Delta T_n$  is relatively small and less than the critical undercooling  $\Delta T_c$ , required for the nucleation of the  $\alpha$  phase, a layer of  $\alpha$  phase called halo structure forms around the primary  $\beta$  phase, which has been often observed in some non-faceted and faceted off-eutectic alloys.

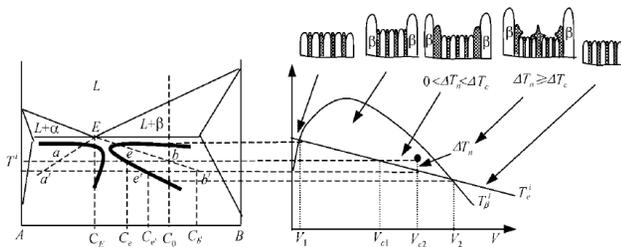


Fig. 4. Coupled zone does not encompass the  $\alpha$  liquidus extension in eutectic systems and interface temperatures of the single  $\alpha$  phase, single  $\beta$  phase and eutectic ( $\alpha+\beta$ ) are calculated by eq. (1) and eq. (3) as a function of growth rates at a given composition  $C_0$  in a hypereutectic alloy [5]

The formation of halo structure is the result of competitive growth between the primary  $\alpha$  phase,  $\beta$  phase and coupled eutectic ( $\alpha+\beta$ ). If the undercooling is more than the critical undercooling ( $\Delta T_n > \Delta T_c$ ),  $\alpha$  phase will nucleate independently not only forming  $\alpha$  halo structure around the primary  $\beta$  phase, but also forming  $\alpha$  dendrites between the primary  $\beta$  phases. Simultaneously,  $\alpha$  phase will grow in preference to coupled eutectic ( $\alpha+\beta$ ) shown in the upper right side of figure 4 and  $\alpha$  dendrites can grow larger in size than the refined coupled eutectic ( $\alpha+\beta$ ).

## 5. Discussion and conclusion

The study of eutectic growth characteristic has shown that the shape and size of the eutectic coupled zone is determined by growth features and solidification conditions. For example, to obtain a proper eutectic growth in the eutectic system containing a pair of faceted/non-faceted phases, the growth of non-faceted phase should be suppressed (or promoted), due to the strong growth anisotropy of the faceted phase.

The undercooling range for the coupled eutectic growth enlarges due to the kinetic effect. The kinetic effect is dependent not only on the growth rate, but also on the type of phase diagram. As the crystallization temperature range of eutectic phases at the eutectic composition decreases, the kinetic effect is enhanced.

The significant difference in linear kinetic coefficient of non-faceted and a faceted phase results in a remarkable difference in kinetic undercooling that plays an important role in influencing the shape of the coupled zone in rapid solidification. To maintain the coupled growth of eutectic phase, the solute undercooling of the faceted phase is weakened in comparison with that of the non-faceted phase by shifting the eutectic composition to the faceted phase side and thus leading to the formation of a skewed locus of eutectic composition in rapid processing. The symmetrical coupled zone with a non-faceted/non-faceted reaction can also be well clarified when their comparable contribution in kinetic undercooling are taken into account.

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