

# Solidification with back-diffusion of irregular eutectics

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## Abstract

The definition of the  $\alpha$  - parameter back-diffusion has been introduced in the work. The alternative models of solidification were described taking into consideration back-diffusion process. The possibility of using those models for eutectic alloys solidification is worthy of interest.

**Keywords:** Solidification; Back-diffusion; Eutectic

## 1. Introduction

Most of the eutectic alloys of practical interest (Fe–C, Al–Si) are irregular eutectics. These and other irregular faceted-nofaceted (f–nf) eutectics are widely employed in industry and are of greater practical importance than the regular eutectics [1].

Directional solidification of binary or pseudo-binary eutectics, may result in regular structures of fibrous or lamellar type. In fibrous growth, one of the phases grows in the form of fibres embedded into a continuous matrix of the other phase, while in the case of lamellar growth, two phases grow cooperatively side by side, in the form of lamellae. When two solid phases  $a$  and  $b$  growing from a liquid of eutectic composition  $\Delta E$ , the average undercooling  $\Delta T$  at the interface results from three contributions:

$$\Delta T = T_E - T_L = \Delta T_c + \Delta T_r + \Delta T_k \quad (1)$$

where  $\Delta T$  is the average interface undercooling,  $T_E$  is the eutectic temperature and  $T_L$  is the local interface temperature, and  $\Delta T_c$ ,  $\Delta T_r$ ,  $\Delta T_k$  are the average undercooling, chemical, capillary and kinetic undercooling, respectively [2].

During the solidification of a dendritic alloy, the solute rejected at the solid/liquid interface is redistributed, by mass diffusion or convection. This process, referred to as microsegregation, controls the composition of the microstructure and the fraction of eutectic or other phases that form. The closed-form, limiting models are the lever rule (complete mixing in the solid and liquid phases) and the Gulliver-Scheil equation (complete mixing in the liquid no diffusion in the solid. When the microstructure can be characterized by a fixed length scale (usually a secondary arm spacing), modifications of the Gulliver-Scheil equation that account for finite-rate diffusion in the solid phase (so called back-diffusion) have been presented in the literature [3].

## 2. The classical Brody and Flemings model

The definition for the  $\alpha$  - back-diffusion parameter worked out by Brody and Flemings is introduced in the analysis [4].  $D_s$  - coefficient of diffusion in the solid,  $t_f$  - local solidification time and  $L$  - half the cell or dendrite spacing all together are deciding on a value of the  $\alpha$  - parameter.

$$\alpha = \frac{D_s t_f}{L^2} \quad (2)$$

The  $\alpha$  - parameter defined by equation (2) can be considered as the ratio of local solidification time  $t_f$  and diffusion time  $t_d$  necessary to ensure the homogeneity of the solid. Taking into account that diffusion time is expressed as a function of diffusion distance:

$t_d = L^2/D_s$ , the back-diffusion parameter  $\alpha$ , equation (2), now becomes:

$$\alpha = \frac{t_f}{t_d} \quad (3)$$

The situation at the liquid–primary solid phase with back-diffusion is depicted schematically in Fig. 1.

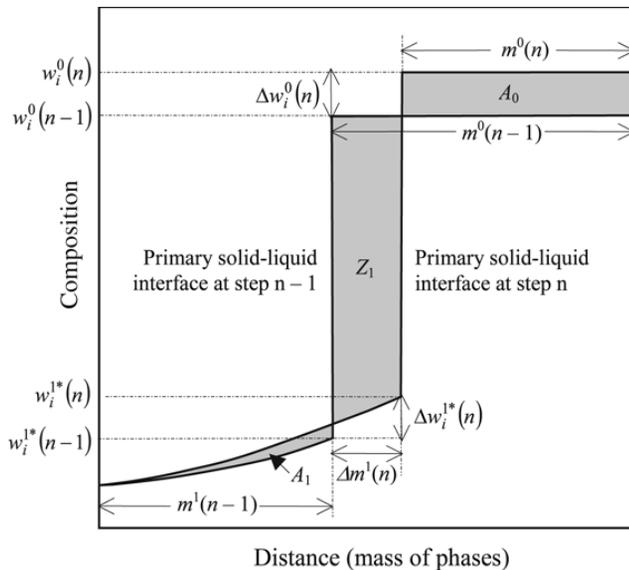


Fig. 1. Schematic illustration of mass distribution of element  $i$  with back-diffusion between the primary solid phase ( $\varphi = 1$ ) and the liquid phase ( $\varphi = 0$ ). The  $x$  axis is distance but the scale is factorized to be linear with mass of phases, where:

$m$  - mass of the system,

$m_i$  - mass of element  $i$  in the system,

$m^\varphi$  - mass of phase  $\varphi$  in the system,

$w_i$  - nominal composition or mass fraction of element  $i$  in the system ( $w_i = m_i/m$ ),

$w_i^\varphi$  - mass fraction of element  $i$  in phase  $\varphi$  ( $w_i^\varphi = m_i^\varphi / m^\varphi$ ),

$\varphi$  - index of phases ( $\varphi = 0$  for the liquid phase,  $\varphi = 1$  for the primary solid phase and other indices are for the secondary solid phases) [5].

The liquid behavior during solidification is described by  $N_L(x;a)$  - solute content. Thus, two extreme cases of solidification are known due to intensity of the solid state diffusion:

- non-equilibrium solidification with complete mixing of solute in the liquid and no diffusion in the solid known as Scheil's model for microsegregation,

$$N_L(x;0) = N_o (-x)^{k-1} \quad (4)$$

where,  $\alpha = 0$  denotes no diffusion of solute into the solid,

- equilibrium solidification where diffusion in solid and liquid are completed at each stage of directional solidification (cellular or dendritic)

$$N_L(x;1) = N_o (+kx - x)^{k-1} \quad (5)$$

where,  $\alpha = 1$  denotes complete diffusion of solute into the solid [4].

### 3. Some alternative growth models

It is worthwhile to describe some alternative models to provide a point of comparison.

#### 3.1. The Voller and Beckermann model

The effect of coarsening can be accounted for in a conventional microsegregation model (*i.e.*, without coarsening) by an additional back-diffusion term. This results in a net diffusion process characterized by the following back-diffusion parameter:

$$a^+ = a + a^c \quad (6)$$

where  $a^c$  is an additive enhancement to the standard Fourier number  $a$  that accounts for the back-diffusion like contribution from coarsening. Equation (6) is important because the effect of coarsening can be included in any microsegregation model by simply replacing the Fourier number  $a$  with the parameter  $a^+$ . Across a wide range of solidification conditions, when the coarsening process goes as  $t^{1/3}$ , the additive enhancement of the Fourier number is constant, taking a value close to  $a^c = 0,1$  [6].

#### 3.2. The Ohnaka model

The model is based upon the assumption that the solute concentration profile can be described by a quadratic expression.

If a parabolic growth law is assumed for the solid phase then:

$$2\alpha = \frac{1}{1 + \frac{LdL}{GDdt}} \quad (7)$$

$$2L \frac{dL}{dt} = \frac{C/2}{t_f} \quad (8)$$

where  $t_f$  is the solidification time and  $\lambda$  is the dendrite arm spacing, which corresponds to twice the size of the model system.

Rewriting the equation, one obtains after inserting Eq. (8) into Eq.(7):

$$2\alpha = \frac{\beta}{1+\beta} \quad (9)$$

where:

$$\beta = \frac{8GDt_f}{\lambda^2} \quad (10)$$

where  $D$  is the solute mass diffusivity and  $G$  is a geometrical parameter in Ohnaka's model [5].

Equation for Ohnaka model is:

$$\frac{C_s}{C_o} = k \left[ 1 - f \left( 1 - \beta k \right)^{\alpha} \right] \quad (11)$$

### 3.3. The Wang–Beckermann model

The back-diffusion given Wang and Beckermann [7] introduce an alternative representation that, under the assumption of a parabolic growth rate, leads to a specific microsegregation model.

If the solid growth is parabolic and a quadratic solute profile ( $n = 2$ ) is assumed the back-diffusion form can be used in a mass balance to arrive at the following microsegregation model:

$$\frac{C_s}{C_o} = \frac{6\alpha \left( 1 - f \right)^{\alpha+1}}{f^{6\alpha}} \int_0^f \varphi^{6\alpha-1} \left( 1 - \varphi \right)^{\alpha+6\alpha\beta} d\varphi \quad (12)$$

For a given value of solid fraction  $f$ , this model can be readily evaluated using a mathematical analysis package [7].

### 3.4. The Nastac–Stefanescu model

Under the assumption that the solid interface concentration,  $C_s^i$  varies slowly with the diffusion time so that a quasi-steady state is rapidly achieved Nastac and Stefanescu arrive at analytical model for microsegregation. This model can account for mass diffusion in both the solid and liquid and can also deal with an arbitrary prescription of the solid growth rate; the model performs well when  $\alpha > 1$ . Under the restrictions of complete liquid diffusion and a parabolic growth rate the general model of Nastac and Stefanescu reduces to:

$$\frac{C_s}{C_o} = k \left[ 1 - \frac{\left( 1 - k \right) f}{1 - kI} \right]^{-1} \quad (13)$$

where:

$$I = \frac{2f}{n^2} \sum_{n=1}^{\infty} \frac{1}{\left( 1 - 0,5 \right)^n} \exp \left[ - \left( 0,5 \right)^n \alpha \right] \quad (14)$$

can be readily evaluated using a mathematical analysis package [7,10].

### 3.5. Voller's specific microsegregation model for parabolic growth

In the case of parabolic solid growth:

$$\beta = \frac{\gamma\alpha}{\alpha + \gamma \left( f / d\tau \right)} \quad (15)$$

With  $f = \sqrt{\tau}$ ,  $f df / dt = 0,5$  becomes

$$\beta = \frac{2\gamma\alpha}{2\alpha + \gamma} \quad (16)$$

If a quadratic solid solute profile,  $C_s = a\zeta^2 + b$ , with constant coefficients is assumed, the rate of change  $\delta C_s = \delta\tau$  will be constant at each point in the solid, and the integration parameter will take the value  $\gamma = 1$ . Comparison with the exact analytical treatment of the parabolic growth, however, shows that this model and its close relative proposed by Clyne and Kurz perform poorly at low  $k$  and  $\alpha$ . A more reasonable model for  $\gamma$ , which is a function of both the Fourier number  $\alpha$  and partition coefficient  $k$  is:

$$\gamma = \frac{A\alpha k}{A\alpha k + 1} \quad (17)$$

where a crude fit with the analytical solution indicates that the constant  $A \sim 4$ . Note that the model in Eq. (17) restricts  $0 \leq \gamma \leq 1$  and matches the required limit values i.e.  $\gamma \rightarrow 1$  as  $\alpha \rightarrow \infty$ , and  $\gamma \rightarrow 0$  as  $\alpha \rightarrow 0$ . The dependence on  $k$  in this model, which is somewhat arbitrary, is driven by the observation that the analytical solution, indicates an increase in the value of  $k$  has the same effect as increasing the Fourier number  $\alpha$  [7].

### 3.6. The Wolczyński model

Conservation of solute within the volume element, which can be created in oriented cellular or dendritic morphology is:

$$d \left( -x \overline{N_L} \right) + d \left( \overline{N_s} \right) = 0 \quad (18)$$

The amount of solute, which leaves the liquid, is:

$$d \left( -x \overline{N_L} \right) = \left( -x \right) d \overline{N_L} - \overline{N_L} dx \quad (19)$$

and that, which causes the growth of crystal is:

$$d \left( \overline{N_s} \right) = \overline{N_s} dx + x d \overline{N_s} \quad (20)$$

The product  $\overline{N_s} dx$  determines an amount of solute within currently solidifying layer  $dx$  and  $x d \overline{N_s}$  expresses an amount of the solute within the  $x$  - solid due to back-diffusion.

The following assumption is introduced into the present model:

$$\overline{N_s} dx + x d \overline{N_s} = \overline{N_s} dx + \alpha \left( x \right) d \overline{N_s} \quad (21)$$

with  $\overline{N_s} \geq \overline{N_s}$  and  $\alpha \left( x \right) \geq 1$

