

# Mathematical and numerical model of directional solidification including initial and terminal transients of the process

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## Abstract

The, developed in this study, simple model and numerical solution of diffusion growth of the solid phase under the conditions of directional solidification allow for the effect of constituent diffusion in both liquid and solid phase and assume the process run in which (like in reality) the preset parameter is the velocity of sample (pulling velocity) at a preset temperature gradient. The solid/liquid interface velocity is not the process parameter (like it is in numerous other solutions proposed so far) but a function of this process. The effect of convection outside the diffusion layer has been included in mass balance under the assumption that in the zone of convection the mixing is complete. The above assumptions enabled solving the kinetics of growth of the solid phase (along with the diffusion field in solid and liquid phase) under the conditions of diffusion well reflecting the process run starting with the initial transient state, going through the steady state period in central part of the casting, and ending in a terminal transient state. In the numerical solution obtained by the finite difference method with variable grid dimensions, the error of the mass control balance over the whole process range was 1 - 2 %.

**Keywords:** Directional solidification, Modelling, Al-Cu

## 1. Introduction

Numerous cases exist when modelling of directional solidification uses the effect of constituent diffusion only, while other phenomena, e.g. convection, are treated in a very restrictive manner. A typical approach to the problem of diffusion growth [1] is based on the already well-established steady state of the process in starting and central part of the sample (casting) placed in an apparatus for directional solidification. Smith et al. [2], Favier [3] extended further the solution of the problem of diffusion growth including also the terminal part of the casting where, due to the effect of boundary surfaces, the process is of a strongly transient character. However, the solutions mentioned above neglect the effect of constituent diffusion in solid phase. Other solutions (Brody and Flemings [4]) consider changes in

boundary conditions, e.g. by introducing limited diffusion in solid with complete diffusion in liquid, allowing for the presence of diffusion layer in liquid (Kurz and Fisher [5]), for the diffusion in solid phase and full mixing in liquid phase (Himemiya and Umeda [6]), and for the diffusion in both liquid and solid phase, though the latter solution, because of boundary conditions, is valid only in micro-scale (Nastac and Stefanescu [7]).

The model proposed by Karma et al. [8] (allowing for the effect of convection) was basis for experimental determination of the diffusion coefficient in the liquid ( $D_L$ ) Pb-Sb2%wt alloy done by Shin et al. [9].

Ellingsen et al. [10], Du, Eskin, Katgerman [11] used CALPHAD software coupled with ThermoCalc software and the, developed by Dore et al. [11] mapping technique to determine solidification path in complex alloys. It seems, however, that there

are still some problems to consider in the directional solidification of two-component alloys, which can be used in further studies of complex systems.

The, developed in this study, simple model and numerical solution of diffusion growth of the solid phase under the conditions of directional solidification allow for the effect of constituent diffusion in both liquid and solid phase and assume the process run in which (like in reality) the preset parameter is the velocity of sample (pulling velocity up) at a preset temperature gradient. The solid/liquid interface (S/L) velocity ( $u_{S/L}$ ) is not treated as a process parameter (like it is done in numerous other solutions proposed so far) but a result of non stationary heat and mass transfer during investigated periods of directional solidification. The effect of convection onto mass transport outside the diffusion layer has been included into a mass balance under the assumption that the mixing is complete in the zone of convection. It has been further assumed after Chen [12] and Shin [9] that the thickness of diffusion layer  $\delta$  is function of a dimensionless parameter  $\Delta$  depending on the sample diameter  $d$ :  $\Delta = \delta \cdot u_p \cdot D_L^{-1/2}$ ;  $\ln(\Delta) = 0.22 - 0.5 \ln(d, \text{mm})$ .

The above assumptions enabled solving the kinetics of growth of the solid phase (along with the diffusion field in solid and liquid phase) under the conditions of diffusion well reflecting the process run starting with the initial transient, going through the steady period in central part of the casting, and ending in a terminal transient. In the numerical solution obtained by the finite difference method with variable grid dimensions, the error of the mass control balance over the whole process range was 1 - 2 %.

## 2. Model of the process

The model assumes that the sample is solidifying with planar solid/liquid (S/L) interface under the conditions resembling those created in a Bridgmann furnace. A unidirectional heat flow along the sample moving at a preset pulling velocity up and the presence of a temperature gradient in the sample have been adopted in the investigations. The growth of solid phase is said to be caused by the concentration gradient of a constituent on the solidification front of a two-component alloy. The effect of mass diffusion in both liquid and solid phase is considered. A model of the boundary diffusion layer [13] of dimension  $\delta$ , interrelated with the sample diameter  $d$  by a dimensionless parameter  $\Delta$ , has been used.

Within the boundary layer, a transient, limited diffusion, depending on the boundary conditions, is assumed to take place (fig. 1). Outside the boundary layer, there is a bulk liquid, in which the transport by diffusion may be neglected [14]. The concentration in bulk liquid depends on the gradient in diffusion layer (at the bulk liquid contact surface) and on the current volume of this bulk liquid.

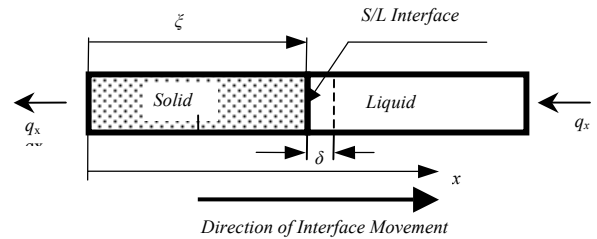


Fig. 1. Scheme of directional solidification

A schematic representation of the phase equilibrium diagram for Al-Cu alloy is shown in Fig. 2.

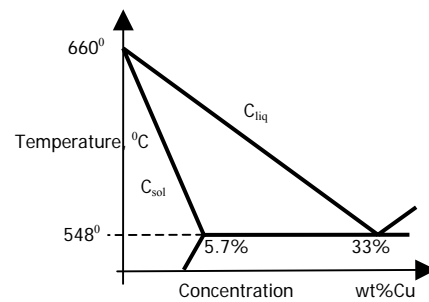


Fig. 2. Schematic representation of the phase equilibrium diagram for Al-Cu alloy

### 2.1. Mathematical model

The concentration field in the examined system is described by the following equations; for the solid state (Fig. 1):

$$\frac{dC_s}{d\tau} = \frac{\partial C_s}{\partial x} \left( \frac{dx}{d\tau} \right) + \frac{\partial C_s}{\partial \tau} \quad (1)$$

where:  $\partial C_s / \partial \tau$  – is taken from the Fick's equation for a fixed interface:

$$\frac{\partial C_s}{\partial \tau} = D_s \frac{\partial^2 C_s}{\partial x^2}$$

where:  $D_s$  – is the coefficient of element diffusion of alloy element in solid.

The term  $dx/d\tau$  in material (substantial) derivative is the velocity of the interface movement  $u_{S/L}$  which can be determined from the following balance equation (Fig. 1 and 2):

$$u_{S/L} (C_L^* - C_S^*) = D_s \frac{\partial C_s}{\partial x} \Big|_{x=\xi} - D_L \frac{\partial C_L}{\partial x} \Big|_{x=\xi} \quad (2)$$

The concentration field in liquid:

$$\frac{dC_L}{d\tau} = \frac{\partial C_L}{\partial x} \left( \frac{dx}{d\tau} \right) + \frac{\partial C_L}{\partial \tau} \quad (3)$$

where  $\left( \frac{dx}{d\tau} \right) = u_{S/L}$

and

$$\frac{\partial C_L}{\partial \tau} = D_L \frac{\partial^2 C_L}{\partial x^2} \quad (4)$$

Boundary conditions:

$$D_s \frac{\partial C_s}{\partial x} \Big|_{x=0} = 0 \quad (5)$$

for  $x = \xi$ ;  $C_s^* = C_{sol}$ ;  $C_L^* = C_{liq}$

$$C_{liq} = (T_0 - T^*) / k_{liq} \quad (6)$$

where  $k_{liq}$  – slope of liquidus line from equilibrium diagram;

$$C_s^* = C_{liq} k_0 \quad (7)$$

where  $-D_L, D_S$  – diffusion coefficient in liquid and in solid

$k_0$  – partition coefficient

$T^*$  – temperature on S/L interface

$T_0$  – melting point of pure base element of alloy

Balance equation at the boundary between the diffusion layer and bulk liquid:

$$X_{BL} \frac{\partial C}{\partial \tau} = D_L \frac{dC_L}{dx} \Big|_{x=\xi+\delta} \quad (8)$$

where  $\xi, \delta, X_{BL}$  – thickness of solid, diffusion layer and bulk liquid

$$\frac{dT^*}{d\tau} = G_T (u_p - u_{S/L}) \quad (9)$$

where  $G_T$  – temperature gradient in the sample

$u_p$  – velocity of the sample (pulling velocity).

## 2.2. Set of difference equations

The equation system was solved by the finite difference method. The differential scheme is shown in Fig. 3.

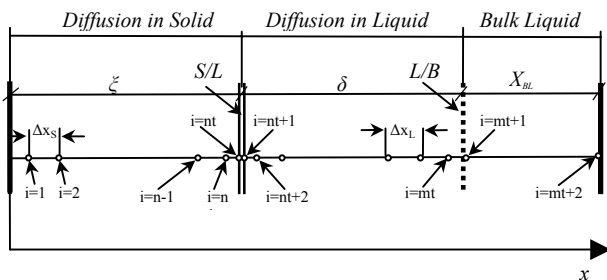


Fig. 3. Differential scheme

In a system of finite differential equations, the calculation nodes were made on the S/L interface and on the mid-dimension of a differential element along with the 3-point derivative.

$$u_{S/L} = (D_L \frac{-8C_{nt+1}^k + 9C_{nt+2}^k - C_{nt+3}^k}{3\Delta x_S} - D_S \frac{C_{n-1}^k - 9C_n^k + 8C_{nt}^k}{3\Delta x_S}) / (C_n^k - C_{nt+2}^k) \quad (10)$$

for  $i \in (l, n-1)$  (Fig. 3):

$$C_i^{k+1} = C_i^k + \frac{x_i}{\xi} \cdot u_{S/L} \cdot \Delta\tau \left( \frac{C_{i+1}^k - C_i^k}{\Delta x_S} \right) + D_{0,S} (C_{i-1}^k - 2C_i^k + C_{i+1}^k) \quad (11)$$

where  $C_i^k$  – concentration of alloy element at  $i$  node for index of time  $k$ ,

$$D_{0,S} = \frac{D_S \cdot \Delta\tau}{\Delta x_S^2} \quad (12)$$

$\Delta\tau, \Delta x$  – time step and grid spacing

$x_i$  – coordinate of differential element

for  $i = n$ :

$$C_n^{k+1} = C_n^k + \frac{x_n}{\xi} \cdot u_{S/L} \cdot \Delta\tau \left( \frac{C_{nt}^k - C_n^k}{\Delta x_S} \right) + \frac{1}{3} D_{0,S} (4C_{n-1}^k - 12C_n^k + 8C_{nt}^k) \quad (13)$$

$$C_{nt}^{k+1} = C_s^* \quad (14)$$

$$C_{nt+1}^{k+1} = C_L^* \quad (15)$$

$$C_{nt+2}^{k+1} = C_{nt+2}^k + \Psi \cdot u_{S/L} \cdot \Delta\tau \left( \frac{C_{nt+3}^k - C_{nt+2}^k}{\Delta x_L} \right) + \frac{1}{3} D_{0,L} (8C_{nt+1}^k - 12C_{nt+2}^k + 4C_{nt+3}^k) \quad (16)$$

where:  $\Psi = \frac{\xi + \delta - x_i}{\delta}$  (influence of decreasing of at the end part of sample)

$$D_{0,L} = \frac{D_L \cdot \Delta\tau}{\Delta x_L^2} \quad (17)$$

if  $X_{BL} > 0$  then  $\psi = 1$

for  $i \in (nt+3, mt-1)$ :

$$C_i^{k+1} = C_i^k + \Psi \cdot u_{S/L} \cdot \Delta\tau \left( \frac{C_{i+1}^k - C_i^k}{\Delta x_L} \right) + D_{0,L} (C_{i+1}^k - 2C_i^k + C_{i+1}^k) \quad (18)$$

for  $i = m_i$ :

$$C_{m_i}^{k+1} = C_{m_i}^k + \Psi \cdot u_{S/L} \cdot \Delta\tau \left( \frac{C_{m_i+1}^k - C_{m_i}^k}{\Delta x_L / 2} \right) + D_{0,L} (C_{m_i-1}^k - 3C_{m_i}^k + 2C_{m_i+1}^k) \quad (19)$$

if  $X_{BL} > 0$  then

$$C_{m_i+1}^{k+1} = C_{m_i+1}^k + \frac{D_L \cdot \Delta\tau (C_{m_i}^k - C_{m_i+1}^k)}{X_{BL} \cdot \Delta x_L / 2} \quad (20)$$

$$C_{m_i+2}^{k+1} = C_{m_i+1}^{k+1} \quad (21)$$

Temperature conditions:

$$T_i^{liq} = T_0 - C_i^k \cdot k_{liq} \quad (22)$$

$$T_{S/L}^{k+1} = T_{S/L}^k - G_T (u_p - u_{S/L}) \Delta\tau \quad (23)$$

$$T_i^{k+1} = T_{S/L}^{k+1} - G_T (x_i - \xi) \quad (24)$$

### 3. The results of modelling

Computer modelling was carried out under the experimental conditions described by Shin et al. [9]. The measurements were taken in a Pb-2%wtSb alloy, and their aim was the determination of constituent diffusivity in liquid phase. To determine this coefficient, the results of the measurements of composition profiles for fraction distances  $f_s = 0.3, 0.5$  and  $0.7$  in samples of  $0.4, 0.8$  and  $1.5$  mm diameters were compared with the results of calculations done on a model developed by Karma et al. [8]. Shin et al. [9] proves that models developed by Smith [2] and Favier [3] deviate from the reality. Allowing for the effect of convection in samples of different diameters, Shin et al. has reached the conclusion that the most reliable model is that developed by Karma, and this model he has used in his calculations and in the determination of the constituent diffusivity in liquid phase.

The results of Shin et al. experiment were presented in Fig. 4 in confrontation with the results of modelling carried out by means of the developed program. The experiment considered the distribution of a constituent (Sb) in frozen sample with the length of the frozen zone amounting to  $50$  mm, which made half length of the whole sample.

The developed model adopts after Shin et al. the values of the convection-related coefficient amounting to  $\Delta = 4, 1$  and  $0.3$  for samples of the diameters of  $0.4, 0.8$  and  $1.5$  mm, respectively. Fig. 4 shows ranges of the  $\delta$ -boundary layer marked in individual samples.

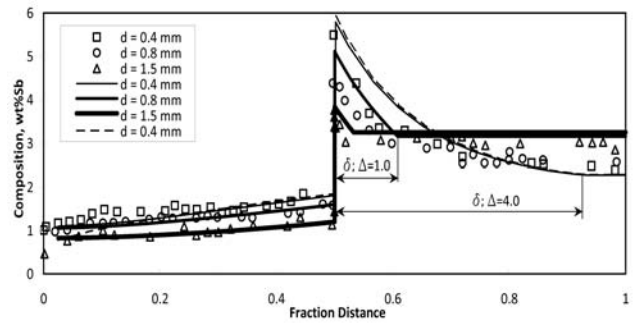


Fig. 4. Measured [9] (symbols) and calculated (lines) compositions profiles in the solid and liquid  $\phi = 0.4, 0.8$  and  $1.5$  mm samples of Pb-2%wtSb. Dashed line:  $D_s = 0$

The results of modelling are similar to the experimental data obtained by Shin et al. These results deviate in the solid phase region from the results obtained on Karma model, not included in the drawing. Only the final outcome (dashed line, Fig. 4) of the calculations made on the developed model under the assumption that the diffusivity in solid phase  $D_s = 0$  (which is corresponding to Karma model) has been shown.

Fig. 5 shows the results of modelling for Al-wt%Cu under the conditions of limited diffusion in liquid phase. In contrast, the Fig. 6 shows concentration field when the boundary layer exists. The dimension of boundary layer is calculated according to [5]  $\delta = DL/u_p$  (according to Chen [12] and Shin [9]  $\Delta = 1$ ). There are visible significant result differences. The summary of differences are shown in Fig. 7 which shows the solute redistribution of Cu in sample at the end of solidification under condition of limited diffusion (solid line) and condition with the boundary layer (dashed line).

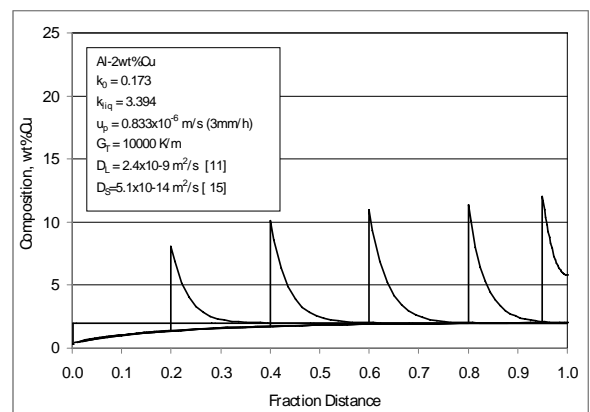


Fig. 5. [15] Solute redistribution of Cu in 100 mm sample under condition with the limited diffusion in liquid phase

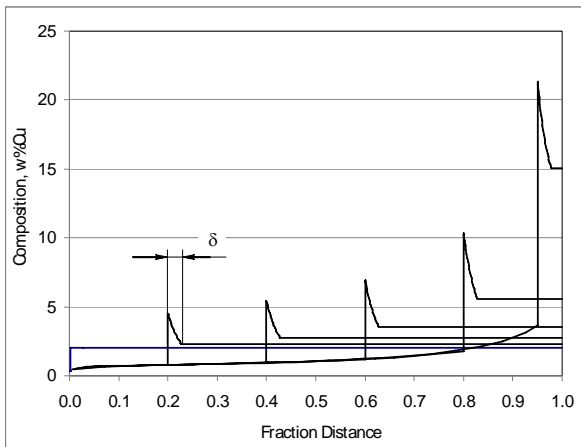


Fig. 6. Solute redistribution of Cu in 100 mm sample under conditions with the boundary layer

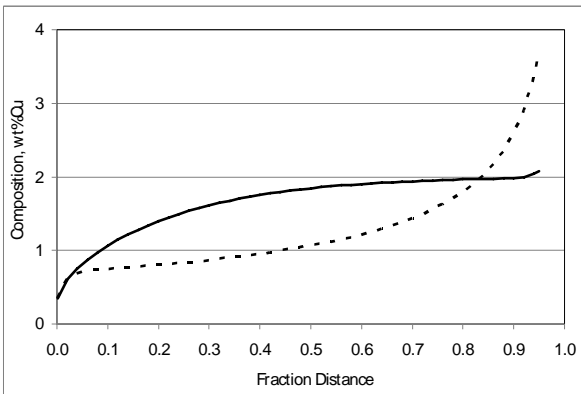


Fig. 7. Solute redistribution of Cu in sample at the end of solidification process. Solid line – condition with limited diffusion; dashed line - condition with the boundary layer

In analytical models with analysis of the experiment done by adjusting method, as an interface velocity the pulling velocity is often adopted. The results of modelling (Fig. 8) show that the difference between the interface velocity and pulling velocity may be quite considerable, especially at higher values of  $u_p$ , and also in the starting and terminal part of the sample.

The developed program enables the constitutional undercooling area to be determined for given process conditions.

The results of modelling indicate absence of the constitutional undercooling area for  $u_p=0.833 \cdot 10^{-6} \text{ m} \cdot \text{s}^{-1}$  (3 mm/h), used in the modelling – Fig. 9a.

With parameters like those specified in the results described above, with exception of the pulling velocity kept at a higher level (threefold,  $u_p=12 \text{ mm/h}$ ), this area appears and increases during the process– Fig. 9b.

The similar influence for possibility of appearing of constitutional undercooling has the temperature gradient in the device (identified as gradient in the sample) – Fig. 9c. Threefold decrease of the temperature gradient in sample causes appearing

of constitutional undercooling, which increases during the process.

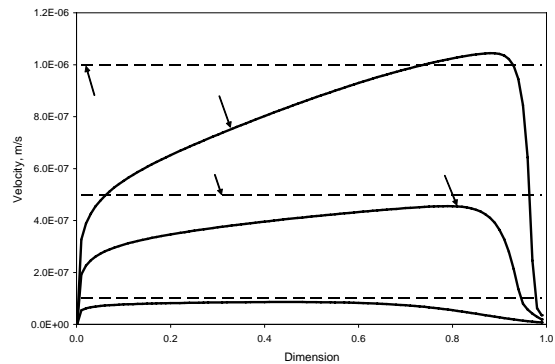


Fig. 8. Velocity of S/L interface (dashed lines) for three pulling sample velocities (solid lines)

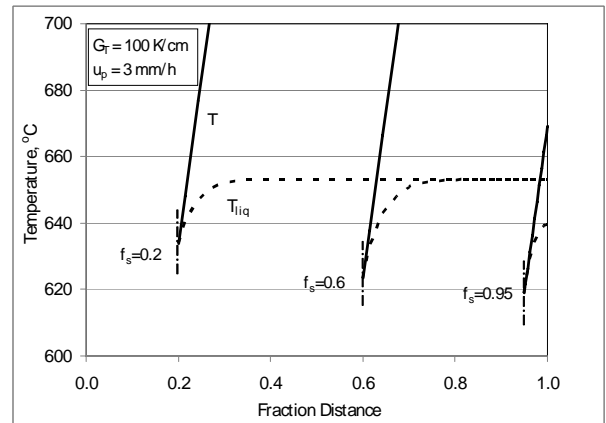


Fig. 9a. The temperature (T) and liquidus temperature ( $T_{liq}$ ) redistribution for the base process parameters

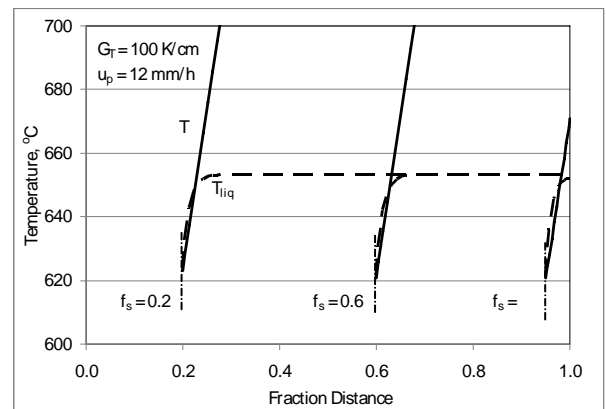


Fig. 9b. The temperature (T) and liquidus temperature ( $T_{liq}$ ) redistribution for the higher pulling velocity - constitutional undercooling during solidification process

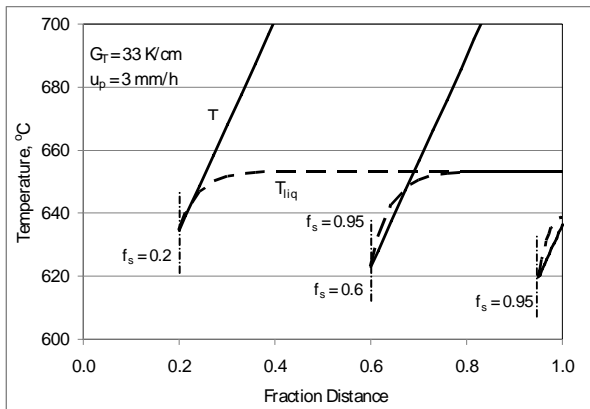


Fig. 9c. Temperature ( $T$ ) and liquidus temperature ( $T_{liq}$ ) redistribution for the lower temperature gradient - constitutional undercooling during solidification process

## 4. Conclusions

A mathematical model and a computer program 1D have been developed for macromodelling of the process run in a device for directional solidification. The results of modelling in respect of the solute redistribution are similar to the results of the experiments available in literature. Allowing in the model for limited diffusion in solid along with the limited diffusion and bulk liquid in liquid phase makes the process more representative and creates the possibility of a reliable determination of the diffusivity in solid phase.

As a result of modelling, an important difference was noted between the pulling velocity  $u_p$  (velocity of sample) and the real velocity of the  $S/L$  interface. The difference is increasing with the growing value of  $u_p$ . It also increases in the starting and terminal area of the sample.

With higher values of  $u_p$  or lower values of temperature gradient, the constitutional undercooling area can be formed, its distribution is changing in time according to process conditions.

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