MODELLING OF PHASE TRANSFORMATIONS IN HEAT TREATMENT PROCESSES

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Abstract. The two-domain approach and the phase-field approach, the two distinct physical models, for simulation of phase transformations in heat treatment processes are presented. Special attention is paid for linking data from a thermodynamic database to the physical models. The general procedure for linking thermodynamic data in the two-domain approach is presented. The interpolation by the radial basis functions of the thermodynamic data in the phase-field model is proposed. The physical models are applied to homogenisation of aluminum alloys. The JMatPro software for aluminium alloys is used as the thermodynamic database. The dissolution kinetics of stoichiometric and nonstoichiometric primary particles in binary and multicomponent aluminium systems is estimated. The isothermal diffusion-controlled dissolutions of the θ phase, S phase and Mg₂Si phase in aluminium phase in Al-Cu, Al-Cu-Mg and Al-Mg-Si systems are computed, respectively. A comparison of the numerical results computed by the physical models are in excellent agreement. The very nice agreement between the numerical results computed by the phase-field model and previously derived the Vermolen model for the dissolution of multicomponent particles in homogenisation of aluminium alloys is demonstrated.

1 INTRODUCTION

Modelling of phase transformations in the solid state is very important for understanding and optimisation of heat treatment processes. The heat treatment processes in alloy production chain are time and energy consuming, therefore their optimisation is an important task in scientific as well as technical communities. Mathematical modelling of the phase transformations has begun more than thirty years ago and still today, it represents an open topic in materials science and engineering [1].
The phase transformations in the solid state are between two extremes, known in literature as the diffusion-controlled (DC) or the interface-controlled (IC) modes. In the DC mode, the main assumption is that the interface reaction proceeds sufficiently fast with respect to the long-distance diffusion in the phases. Therefore, the interface concentrations are determined through thermodynamic equilibrium. If the interface reaction proceeds slower than the long-distance diffusion in the phases, the phase transformations are IC. The transformation is in a mixed-mode character if both processes influence on the transformation kinetics. The transition from the one to the other mode depends upon the relative magnitudes of the solute diffusion and the interface mobility. The scope of this paper is to demonstrate the linking of the data from a thermodynamic database to the physical models. Therefore, the analysis is confined to DC phase transformations only.

Modelling of the phase transformations falls within the wide spectrum of moving boundary problems. A lot of numerical procedures for solving moving boundary problems exist [2]. Numerical procedures are divided into two major groups: one domain approaches and two domain approaches.

The two separate physical models, the two-domain approach (TDA) and the phase-field approach or model (PFM), for phase transformations are analysed. In the TDA, the fields are solved separately for the domains occupied by the different phases. The major problem is formulation of boundary conditions on (usually) geometrically complex moving boundaries. The positions of boundaries are difficult to track in a multidimensional topology.

The PFM has been developed as a basis for powerful computational modelling of complex phase-change systems ([3],[4]). The PFM belongs to one-domain approaches, where the same governing equations hold in the whole domain. The PFM introduces nonconserved phase-field variable that is continuous across the interphase boundary (i.e. interface). The behavior of this variable is governed by a phase-field equation that is coupled to the equations for heat and solute transport. Interfaces are described by smooth, however highly localized changes of this variable between the fixed values that represent bulk phases. The problem of tracking of the moving boundary is thus transformed into a problem of proper presentation of the large gradients of the phase-field variable near the moving interface. A great advantage of the one-domain approaches is the possibility to use the fixed (however sufficiently dense) mesh numerical schemes throughout computation.

This paper is organized as follows. Some general characteristics of models are written in Second section. The TDA for phase transformations in multicomponent alloys is demonstrated in Third section. Elaborated is particularly the connection with a thermodynamic database. The PFM is presented in Fourth section. Special attention for coupling of the thermodynamic databases with the PFM. The numerical results of isothermal DC dissolution of primary particles in aluminium alloys during homogenisation computed by the both models are presented in Fifth section. The dissolutions of the $\theta$ phase, S phase and Mg$_2$Si phase in Al-Cu, Al-Cu-Mg and Al-Mg-Si systems are estimated. The conclusions and future development are exposed at the end.
2 GENERAL CHARACTERISTICS OF THE PHYSICAL MODELS

Consider a domain $\Omega$ with boundary $\Gamma$, filled with a multicomponent alloy with $M$ different components and two different phases $\alpha$ and $\beta$. The system is in thermal and mechanical equilibrium. The purpose of this paper to calculate the concentrations of the components in different phases as a function of position $\vec{p}$ and time $t$, throughout a change of temperature $T$. The $M-1$ independent concentration fields have to be computed in the whole domain, because the following constraint holds

$$\sum_{m=1}^{M} c_{\eta}^m = 1$$  \hspace{1cm} (1)

where $c_{\eta}^m$ is the concentration of component $m$ in phase $\eta$ (wt%), $\eta = \alpha, \beta$. In this paper, superscript denotes a component and subscript a phase, respectively. The concentration vector of alloying elements in phase $\eta$ is denoted in bold as $\vec{c}_{\eta} = [c_1^\alpha c_2^\alpha \ldots c_{(M-1)}^\alpha]^T$. All boundary conditions are assumed of the Neumann type, that is no diffusion across the system boundary

$$\nabla \vec{c}_{\eta} \cdot \vec{n}_\Gamma = 0; \ m = 1,2,\ldots,M-1 \equiv \nabla \vec{c}_{\eta} \cdot \vec{n}_d = 0$$  \hspace{1cm} (2)

where $\vec{n}_\Gamma$ is normal vector on the exterior boundary.

3 THE TWO-DOMAIN APPROACH

The description of TDA is divided into three different segments: the solute diffusion equation in the bulk phase, the thermodynamic equilibrium at the interface and the mass conservation at the interface. Summary and drawbacks of the present approach are written at the end.

3.1 Solute diffusion equation in the bulk phase

The solute diffusion equation of alloying element $m$ ($m = 1,2,\ldots,M-1$) in the bulk phase $\eta$ is obtained from the classical mass conservation law as

$$\frac{\partial c_{\eta}^m}{\partial t} = -\nabla \cdot \vec{J}_{\eta}^m; m = 1,2,\ldots,M-1, \eta = \alpha, \beta$$  \hspace{1cm} (3)

where $c_{\eta}^m$ is the concentration of component $m$ and $\vec{J}_{\eta}^m$ is the multidimensional mass flux of component $m$ in the phase $\eta$ ($\eta = \alpha, \beta$). The vector of mass fluxes in the phase $\eta$ is denoted as $\vec{J}_\eta = [\vec{J}_1^\eta \vec{J}_2^\eta \ldots \vec{J}_{(M-1)}^\eta]^T$. The vector of mass fluxes represent a product of the interdiffusion coefficient matrix and the vector of concentration gradients of alloying elements in general:

$$\vec{J}_\eta = -\eta \tilde{D} \nabla \vec{c}_\eta$$  \hspace{1cm} (4)
where the isotropic interdiffusion coefficient matrix and the vector of concentration gradients are

$$
\eta \tilde{D} = \begin{bmatrix}
\tilde{D}_{11} & \tilde{D}_{12} & \ldots & \tilde{D}_{1(M-1)} \\
\tilde{D}_{21} & \tilde{D}_{22} & \ldots & \tilde{D}_{2(M-1)} \\
\vdots & \vdots & \ddots & \vdots \\
\tilde{D}_{(M-1)1} & \tilde{D}_{(M-1)2} & \ldots & \tilde{D}_{(M-1)(M-1)}
\end{bmatrix}, \quad \nabla c_\eta = \begin{bmatrix}
\nabla c^1_\eta \\
\nabla c^2_\eta \\
\vdots \\
\nabla c^{(M-1)}_\eta
\end{bmatrix}
$$

The off-diagonal entries $\eta \tilde{D}_{nm}$ ($i \neq j$) of interdiffusion coefficient matrix, also referred to as the cross-terms, are the measure for the interaction between consecutive alloying elements in the phase $\eta$. When $\eta \tilde{D}_{nm} < 0$, then alloying element $m$ deteriorates diffusion of the element $n$ in the phase $\eta$, whereas $\eta \tilde{D}_{nm} > 0$ implies that the element $m$ facilitates the diffusion of the element $n$ in the phase $\eta$.

Here, we use the assumption that the diffusion of each alloying element is independent on the presence of any alloying element ($\eta \tilde{D}_{nm} = 0, n \neq m$). There are two reasons for using this assumption: The first one is that the cross-diffusion coefficients $\eta \tilde{D}_{nm}$ are much lower than the diagonal diffusion coefficients $\eta \tilde{D}_{mm}$, the second one is that the cross-diffusion coefficients of interdiffusion coefficient matrix in phases can not be easily obtained from literature. The DICTRA software with different mobility databases represents one of the ways for obtaining the interdiffusion coefficient matrices for various phases. In what follows, the diffusion coefficients are denoted without tilde, and the subscript $mm$ is replaced with $m$.

If we take into account the above mentioned assumption, the mass fluxes of alloying elements do not dependent of the concentration gradients of other elements

$$\vec{J}_\eta^m = -\eta D_m \nabla c^m_\eta \quad (6)$$

and the solute diffusion equation of alloying element $m$ becomes

$$\frac{\partial c^m_\eta}{\partial t} = \nabla \cdot \left( \eta D_m \nabla c^m_\eta \right) \quad (7)$$

If we use another simplification for diffusion coefficients to be constant throughout the domain, the final solute diffusion equation of component $m$ in the phase $\eta$ is

$$\frac{\partial c^m_\eta}{\partial t} = \eta D_m \Delta c^m_\eta \quad (8)$$

### 3.2 Thermodynamic equilibrium at the interface

In the DC transformation mode, the thermodynamic variables at the interface in both phases are in thermodynamic equilibrium. Firstly, we will see the number of degrees of freedom, which determines the number of variables that can be obtained from thermodynamics. The number of degrees of freedom $F$ in a thermodynamic system is calculated as
a function of the number of components $M$ and the number of phases $P$ from the Gibbs phase rule

$$F = M - P + 2$$

(9)

Solid-solid phase transformations in heat treatment processes usually occur at constant pressure, therefore the number of degrees of freedom is lowered by one, $F = M - P + 1$. In this paper, we are confined to analyzing transformations between two phases ($P = 2$). Consequently the number of degrees of freedom is

$$F = M - 1$$

(10)

Therefore, the number of degrees of freedom in isobaric two-phase systems is $F = 1$ in a binary system ($M = 2$), $F = 2$ in a ternary system ($M = 3$), $F = 3$ in quaternary system ($M = 4$), etc.

The unknown thermodynamic variables at the interface of phases $\alpha$ and $\beta$ are

$$T, c^m_\alpha, c^m_\beta ; \ m = 1, 2, \ldots, M - 1$$

(11)

where $c^m_\eta$ is the concentration of component $m$ in phase $\eta$ on the interface. The number of unknown thermodynamic variables at the interface is $1 + 2(M - 1)$.

The phase transformations in the solid state occur much slower than temperature changes at the micro scale and usually the temperature profile is known from the macroscopic level

$$T = T(t)$$

(12)

Thermal and mechanical equilibrium are assumed earlier. From chemical equilibrium one can get $M$ independent equations

$$\mu^m_\alpha (T, c^m_\alpha) = \mu^m_\beta (T, c^m_\beta) ; \ m = 1, 2, \ldots, M$$

(13)

where $\mu^m_\eta$ is the chemical potential of component $m$ in phase $\eta$. Eq.(13) holds for each of components $M$ in the system. These equations represent the condition that has to be satisfied in order for any two phases to coexist in thermodynamic equilibrium. They represent equilibrium conditions in a multicomponent two phase system [5]. If we use a thermodynamic database, $M$ equations can be chosen as

$$f_\alpha (T, c^m_\alpha) = 0 ;$$
$$f_\beta (T, c^m_\beta) = 0 ;$$
$$F_n (T, c^m_\alpha, c^m_\beta) = 0 ; \ n = 1, 2, \ldots, M - 2$$

(14)

where $f_n$ is the mathematical description of the $M$-dimensional surface of phase $\eta$. The mathematical description of tie-lines can be written as independent $M - 2$ equations for $F_n$. 

5
Stoichiometric phase. An intermetallic compound with fixed concentration is known as a stoichiometric phase. Numerous stoichiometric phases are present in ferrous and non-ferrous alloys. The conditions of the thermodynamic equilibrium at the interface between a stoichiometric and ‘normal’ phase are different [6]. Therefore, it will be analysed here separately. We denote the chemical species by $S_m$, $m = 1, 2, \ldots, M$ and the stoichiometry compound of the particle by $(S_1)_{n_1} (S_2)_{n_2} \ldots (S_M)_{n_M}$ (for example Al$_2$CuMg), where the numbers $n_1, n_2 \ldots n_M$ are stoichiometric constants.

The number of degrees of freedom is equal to $F = M - 1$, the same as in the previous case. The unknown thermodynamic variables at the interface are

$$T, c_{\alpha I}^m : \quad m = 1, 2, \ldots, M - 1$$

(15)

The number of unknown thermodynamic variables is $1 + (M - 1)$, because the concentrations of the stoichiometric $\beta$ phase is known ($c_\beta = c_\beta^\beta$). The number of unknown thermodynamic variables minus the number of degrees of freedom is one; Only one law or equation can be obtained from thermodynamic equilibrium, consequently. Hence, the most general condition for the mathematical model is

$$f_\alpha (T, c_{\alpha I}) = 0$$

(16)

which guarantees that the interface concentrations in the phase $\alpha$ are on the $M$-dimensional surface $\alpha$.

If we assume that the phase $\alpha$ is dilute solution, an hyperbolic relation between the interfacial concentrations is valid

$$\prod_{m = 1}^{M-1} \bar{c}_{\alpha I}^{mm} = K(T)$$

(17)

where $\bar{c}_{\alpha I}^{mm}$ is the molar concentration of component $m$ in the $\alpha$ phase at the interface (molar quantities are denoted by overbar in this text) and the factor $K$ is referred to as the solubility product. The solubility product depends on temperature $T$ according to an Arrhenius relationship. The Vermolen model [7, 8] is based on this kind of the interface condition Eq.(17).

3.3 Mass conservation at the interface

The mass conservation of alloying element $m$ at the interface can be written as

$$\left( c_{\beta I}^m - c_{\alpha I}^m \right) \frac{\partial l}{\partial t} = \vec{J}_{\alpha I}^m \cdot \vec{n}_{\alpha \beta} - \vec{J}_{\beta I}^m \cdot \vec{n}_{\alpha \beta}$$

(18)

where $\partial l/\partial t$ is the interface velocity, $l$ is the interface position, $\vec{n}_{\alpha \beta}$ is the vector normal from the phase $\alpha$ into the phase $\beta$ on the interface, $\vec{J}_{\alpha I}^m$ is the interface mass flux of
component \( m \) in phase \( \eta \). If phase \( \beta \) is stoichiometric, the mass conservation at the interface becomes

\[
\left( c_{\beta m}^m - c_{\alpha m}^m \right) \frac{\partial \ell}{\partial t} = \vec{J}_{\alpha I}^m \cdot \vec{n}_{\alpha \beta}
\]

(19)

In binary alloys, this equation is known as the classical Stefan problem [9]. In multicomponent alloys, the number of species to be conserved is \( M - 1 \) and in literature this set of equations is known as a vector-valued Stefan problem. The one-phase classical or vector-valued Stefan problem has to be solved for binary or multicomponent alloys respectively, if phase \( \beta \) is stoichiometric.

The number of mass conservation equations at the interface is \( M - 1 \) and the only one additional unknown value, the interface velocity \( \partial \ell / \partial t \), is included. Because of that, the system of equations from the mass conservation law at the interface is overdetermined by \( M - 2 \).

3.4 Summary

The total number of unknowns at the interface between two phases in thermodynamic equilibrium is \( 2(M - 1) + 1 + 1 = 2M \), which are the thermodynamic variables in Eq.(11) and the interface velocity in Eqs.(18). The mathematical model based on the TDA consists of the following system of equations: the \( M - 1 \) equations for concentration fields of alloying elements in the bulk phases (Eqs.(3)), the \( M \) equations from thermodynamic equilibrium of type Eqs.(13) or Eqs.(14) and the \( M - 1 \) mass conservation equations at the interface (Eqs.(18)) assuming the known temperature profile (Eq.(12)).

The total number of unknowns at the interface between a stoichiometric particle and the matrix is \( (M - 1) + 1 + 1 = M + 1 \), the thermodynamic values in Eq.(15) and the interface velocity in Eqs.(19). For using the TDA for the phase transformations of stoichiometric phase one have to solve: the \( M - 1 \) equations for concentration fields of alloying elements in the matrix (Eqs.(3)), the \( M - 1 \) mass conversation equations at the interface Eqs.(19) and the thermodynamic equation Eq.(16) or Eq.(17) assuming the known temperature profile (Eq.(12)).

The general model based on the TDA with thermodynamic equilibrium at the interface is presented here. The major drawback of the proposed model is formulation of boundary conditions (Eq.(18) or Eq.(19)) on geometrically complex moving boundaries. The interface positions are difficult to track in multidimensional topology. The curvature effect is neglected. Shortly, the model is limited to one-dimensional geometry.

Multidimensional models are necessary for a reasonable understanding and qualitative modelling of the phase transformations during heat treatment processes. The phase-field model, a representative of the one-domain approach, has potential to cope with the above mentioned difficulties.
4 THE PHASE-FIELD APPROACH

The PFM introduces the phase-field variable $\phi (\vec{r}, t)$ which describes the phase-state of the material. In this paper, $\phi = 0$ represents the phase $\alpha$ and $\phi = 1$ represents the particle of the phase $\beta$. Similarly, the concept of the multiphase-field model can be introduced.

The phase-field equation is developed by the free energy functional or the entropy of the system. We present the formulation derived through the free energy functional [3]. The free energy functional $\mathcal{F}$, expressed through the Ginzburg-Landau theory is

$$\mathcal{F} = \int_V \left( \bar{f}(\phi, c, T) + \frac{\varepsilon_\phi^2}{2} |\nabla \phi|^2 \right) dV$$

where $\bar{f}(\phi, c, T)$, $\varepsilon_\phi$, $c$ and $T$ represents the molar free energy density, the energy gradient coefficient, concentrations of alloying elements and temperature, respectively.

For time-dependent situations, the simplest equations that guarantee a decrease in the total free energy with time are postulated as

$$\frac{\partial \phi}{\partial t} = -M_\phi \frac{\partial \mathcal{F}}{\partial \phi}$$

where $M_\phi$ represents the positive mobility coefficient and $\partial \mathcal{F}/\partial \phi$ represents the variational derivative. In mathematician community, Eq.(21) is called the Allen-Cahn equation. By using Eq.(20), Eq.(21) becomes

$$\frac{\partial \phi}{\partial t} = -M_\phi \left( \frac{\partial \bar{f}}{\partial \phi} - \varepsilon_\phi^2 \nabla^2 \phi \right)$$

The free energy density can be constructed in several ways, as well. Various PFMs define the free energy density in different ways. Also, the free energy density of particular phases can be obtained by a thermodynamic database. In proposed PFM, the free energy density $\bar{f}(\phi, c, T)$ is postulated as

$$\bar{f}(\phi, c, T) = p(\phi) \bar{f}_\beta(c, T) + [1 - p(\phi)] \bar{f}_\alpha(c, T) + W g(\phi)$$

where $\bar{f}_\eta(c, T)$ is the molar free energy of phase $\eta$ and $W$ is energy hump between phases. An interpolation $p(\phi)$ and a double-well $g(\phi)$ function in the free energy density are defined by

$$p(\phi) = \phi^3 (6\phi^2 - 15\phi + 10)$$

$$g(\phi) = \phi^2 (1 - \phi)^2$$

Eqs.(24) ensure that $\partial \bar{f}/\partial \phi = 0$, when $\phi = 0$ and $\phi = 1$. The phase-field equation is obtained by inserting Eqs.(23,24) into Eq.(22)

$$\frac{\partial \phi}{\partial t} = M_\phi \left[ \varepsilon_\phi^2 \nabla^2 \phi - 2W(1 - \phi)(1 - 2\phi) - 30\phi^2 (1 - \phi)^2 D_{\beta\alpha} \right]$$
where $D_{\beta\alpha}(c, T)$ is the difference between the free energy of phases, $\bar{f}_{\beta} - \bar{f}_{\alpha}$. This value is known as the driving force for transformation of the phase $\beta$ into the phase $\alpha$. If $D_{\beta\alpha} < 0$ the phase $\beta$ grows from the phase $\alpha$ and if $D_{\beta\alpha} > 0$ the phase $\beta$ dissolves into the phase $\alpha$. For an isothermal phase transformation, the driving force for transformation of the phase $\beta$ into the phase $\alpha$ per mole of the phase $\beta$ is given by

$$D_{\beta\alpha}(c) = \bar{f}_{\beta}(\bar{c}_{\beta}) - \sum_{m=1}^{M-1} \bar{c}_{\beta}^m \left[ \mu_{\alpha}^m(\bar{c}_{\alpha}) - \mu_{\alpha}^M(\bar{c}_{\alpha}) \right]$$  \hspace{1cm} (26)$$

where $\bar{c}_{\beta}^m$ is the molar concentration of component $m$ in the phase $\beta$, $\mu_{\alpha}^m$ is the chemical potential of alloying component $m$ in the phase $\alpha$ and $\mu_{\alpha}^M$ is the chemical potential of the main constitutive element of the same phase.

The phase-field variable $\phi$ in diffusion interface varies in the x normal direction to the interface as

$$\phi(x) = \frac{1}{2} \left[ 1 + \tanh \left( \frac{3x}{\delta} \right) \right]$$  \hspace{1cm} (27)$$

where $\delta$ is a measure of the interface thickness. The model parameters $\epsilon_{\phi}$ and $W$ are determined by the measure of the interface thickness $\delta$ and the interface energy $\sigma$ as

$$\epsilon_{\phi} = \sqrt{\sigma \delta}, \quad W = 18\sigma/\delta$$  \hspace{1cm} (28)$$

respectively. The mobility coefficient $M_{\phi}$ is expressed as $M_{\phi} = \mu^{*}/(\delta \Delta s_{\beta\alpha})$, where $\mu^{*}$ and $\Delta s_{\beta\alpha}$ represent the interface kinetic coefficient and the entropy difference between the phases, respectively.

In the original PFM, the solute diffusion equation for binary alloys was derived [11] from minimisation of the free energy functional. Later, an idea of the averaging method was presented in [12]. A comparison between solute diffusion equations derived from both concepts is explained in the later paper.

The mixture concentration $c$ is introduced as

$$c = \phi c_{\beta} + (1 - \phi) c_{\alpha}$$  \hspace{1cm} (29)$$

The mixture concentration is equal to concentration of phase in bulk phases. At the diffuse interface, the mixture concentration is defined by the concentrations of phases, as is evident from Eq.(29). The one group of PFMs define that the mixture concentration is equal to the concentrations of phases

$$c = c_{\alpha} = c_{\beta}$$  \hspace{1cm} (30)$$

We use the concept of the PFM, where the concentrations of phases are different at the diffuse interface. Therefore, another constraint for defining the concentrations of phases is needed. The constraint for computing the concentrations of phases is that the chemical potentials of components in both phases are equal [13]

$$\mu_{\alpha}^m - \mu_{\alpha}^M = \mu_{\beta}^m - \mu_{\beta}^M; \quad m = 1, \ldots, M - 1$$  \hspace{1cm} (31)$$
This condition is used when the concentrations of phases are not constant.

If the phase $\beta$ is stoichiometric, the concentration of this phase is equal to

$$ c_\beta = c_{\beta}^{st} \quad (32) $$

At the diffuse interface, the concentrations of the alloying elements in the phase $\alpha$ can be computed by

$$ c_\alpha = \frac{c - \phi c_{\beta}^{st}}{1 - \phi} \quad (33) $$

Now, the solute diffusion equation of component $m$, valid over the whole system, is

$$ \frac{\partial c^m}{\partial t} = \nabla \cdot \left( \phi \vec{J}^m_\beta + (1 - \phi) \vec{J}^m_\alpha \right) \quad (34) $$

The concentration flux $\vec{J}^m_\eta$ of component $m$ in phase $\eta$ is written in Eq.(6). The approach is based on the physical statement that the mixture concentration $c$ in an infinitesimally small volume or in the finite control volume of the numerical model can only change by external fluxes over the boundary of this volume. These fluxes are superposed according to the local phase densities, which have to be evaluated on the boundary of the volume.

### 4.1 Interpolation of thermodynamic data by radial basis functions

Thermodynamic data for the physical models are obtained from a thermodynamic database. We do not use a direct link to a database and data have to be prepared for modelling. The chemical potentials of components in the phases as a function of the concentrations are necessary for quantitative computation in the PFM. The data are usually prepared in an discretized or tabular form in the finite number of points $N$

$$ \mu^m_\eta = \mu^m_\eta \left( c^m_\eta \right); \quad n = 1, 2, \ldots, N \quad (35) $$

We promote the interpolation by the radial basis functions in form

$$ \mu^m_\eta = \mu^m_\eta \left( c^m_\eta \right) = \sum_{n=1}^{N} \varsigma^m_\eta,n \psi_n \left( \| c^m_\eta - c^m_{\eta,n} \| \right) \quad (36) $$

where $\psi_n$ and $\varsigma_n$ are the radial basis functions (RBFs) and its coefficients, respectively. The RBFs $\psi_n$ are only functions of the distance between the concentration $c^m_\eta$ and the centers of RBFs $\psi_n$, $c^m_{\eta,n}$. This interpolation enables a very accurate computation of the chemical potential for an arbitrary value of concentration. This interpolation also allows very accurate and easy calculation of derivatives

$$ \frac{\partial \mu^m_\eta}{\partial c^m_\eta} = \sum_{n=1}^{N} \varsigma^m_\eta,n \frac{\partial \psi_n}{\partial c^m_\eta} \quad (37) $$
If the phase $\beta$ is nonstoichiometric, Eqs.(29,31) have to be solved in each node that falls into the diffuse interface. Quick iterative solvers for the system of Eq.(29,31) can be used for precise computation.

The characteristics of proposed interpolation are the following. The interpolation of functions and derivatives is very accurate. The number of the data from a thermodynamic database $N$, can be significantly lower. Temperature can be included in the interpolation. The implementation for multicomponent alloys is straightforward.

The RBFs collocation methods can be used not only for data interpolation but also solving of the phase-change processes [14, 15].

5 DISSOLUTION OF PRIMARY PHASE IN ALUMINIUM ALLOYS

The physical models are written for a general solid state two-phase system. The numerical results are presented for aluminium alloys. The results are focused to solution heat treatment or homogenisation of aluminium alloys. The JMatPro thermodynamic database for aluminium alloys [16] is used in numerical computations. Diffusivity coefficients are obtained from Smithhells Metals Reference Book [17].

The isothermal DC dissolution of primary particles during homogenisation in binary Al-Cu and ternary systems Al-Cu-Mg, Al-Mg-Si is treated. The numerical procedure proposed by Vermolen and Vuik for solution with the front-tracking method (FTM) as a representative of the TDA is used [18]. The FTM and PFM are discretized by the classical second-order finite difference method in space. The FTM is integrated in time using the implicit and the PFM explicit time step scheme. The numerical results computed by both models are presented on the same graphs to demonstrate the possibility of the PFM for quantitative modelling of the transformations in the solid state. Because of the almost coincident results, the representative data are given in a tabular form.

The results are presented for a one-dimensional system with length of 50 $\mu$m. Simulation time is one hour. Initial concentrations in the aluminium phase are assumed constant, because of simplicity. Two equidistant fixed meshes are used for discretization of the PFM. The first mesh is around the interface and the coarser one out of the phase-change region. Distance between neighbouring nodes are for the first mesh $2 \times 10^{-2} \mu$m and for the second $2 \times 10^{-1} \mu$m. The total number of nodes is 701. The measure of the interface thickness is $1.2 \times 10^{-1} \mu$m. The driving force for phase transformations are computed as explained in previous section. The interpolation by radial basis functions is used for accurate computation of the driving force.

The very narrow diffusion interface in the PFM ($1.2 \times 10^{-1} \mu$m) is used for getting sufficiently grid independent results. Therefore, the distance between the neighbouring nodes in the diffuse interface region has to be smaller. Consequently, the time-step length has to be shorter. The number of nodes in the diffusion interface is 6. Also, the interface kinetic coefficient indirectly influences on the time-step length through the positive mobility coefficient $M_\phi$ in Eq.(25).

The interface energy and the interface kinetics coefficient in the PFM have to be es-
The interfacial energy depends on the type of the particle-aluminium phase interface [19]. The interface energy is $5.0 \times 10^{-1} \text{ J/m}^2$ in each computation. The interface energy in the one-dimensional systems does not have a large influence on the transformation kinetics. The interface energy significantly depends on particles shape. The interface kinetics coefficient defines the mode of the phase transformation: diffusion-controlled, interface-controlled or mixed-mode. The interface energy coefficient is not known and it is estimated by numerical experiments. This comparison represents the way for estimating the interface kinetic coefficient in DC phase transformations.

The difference between the numerical results is defined as

$$\Delta = \frac{|V_{\text{FTM}} - V_{\text{PFM}}|}{V_{\text{FTM}}} \times 100\% \quad (38)$$

where $V_{\text{FTM}}$ and $V_{\text{PFM}}$ are values computed by the FTM and the PFM, respectively. The interface position in the PFM is computed implicitly from the phase-field variable, where $\phi = 0.5$. The linear interpolation between neighbouring nodes where $\phi = 0.5$ is used for computing the interface position.

### 5.1 Al-Cu system

Copper is the most common alloying element for aluminium alloys, and a variety of alloys (2xxx series) in which copper is the major addition were developed. The Al-Cu alloy solidifies with eutectic reaction

$$\text{liq} \rightarrow \text{Al} + \theta \quad (39)$$

at 548°C. Copper is solvable up to 5.7 wt% in the aluminium (Al) phase. The Al has a face-centered cubic (FCC) crystal structure and phase $\theta$ has the approximate composition $\text{Al}_2\text{Cu}$ and a complex body-centered tetragonal structure. When the copper content increases, there is a continuous increase in hardness, but strength and especially ductility depend on the shape of the copper-rich $\theta$ phase. The difference is between a spheroidised and evenly distributed particles and a continuous network at the grain boundaries. Dissolved copper produces the highest increase in strength. The network of the copper-rich phase has a strong negative influence on the ductility [20].

The basic purpose of homogenisation of 2xxx aluminium alloys is to dissolve the interdendritic eutectic, the mixture of the phases $\theta$ and Al. In this section, we demonstrate the numerical results of the dissolution of the $\theta$ phase in Al phase, computed by the FTM and PFM. Dissolution of the eutectic phase in Al phase during homogenisation is modelled in [21], where the eutectic phase is treated as a heterogeneous mixture of solid phases.

The initial concentrations of Cu in the Al and $\theta$ phase are constant and equal to 0.5 wt% and 53.0 wt%, respectively. The position of the initial interface is $10\mu\text{m}$. The total number of nodes in the FTM is 402, whereas the number of nodes in the $\theta$ phase and Al phase are 81 and 321, respectively. The number of nodes is determined from the phase...
transformation of the $\text{Al}_3\text{Mg}_2$ phase in Al phase in Al-Mg system, where the diffusion length of Mg in the Al phase ($D_{\text{Mg}}/(\partial l/\partial t)$) is much higher than the diffusion length of Cu in the Al phase. The time-step length in the TDA is $5 \times 10^{-2}$ s. The interface concentrations of Cu in the $\theta$ and Al phase are directly determined from homogenisation temperature by using the JMatPro software.

An modified Powell hybrid algorithm for system of Eqs.(29,31) is used for computation of the concentration of the phases. The interface kinetic coefficient divided by entropy difference between phases is $5.0 \times 10^{-13}$ m$^4$/Js. The time-step length in the PFM is $5 \times 10^{-5}$ s.

The concentration profiles of Cu after one hour of isothermal DC dissolution of the $\theta$ phase in Al phase at three homogenisation temperatures computed by the FTM and PFM are presented in Fig.1. The interface positions during isothermal DC dissolution of the $\theta$ phase in Al phase are presented in Fig.(2). Temperatures 480$^\circ$C, 500$^\circ$C and 520$^\circ$C are chosen as homogenisation temperatures. One can see excellent agreement between the numerical results computed by the models. Evident is a huge influence of homogenisation temperature on the dissolution kinetics.

![Figure 1: The concentration profiles of Cu after one hour of isothermal DC dissolution of the $\theta$ phase in Al phase at three homogenisation temperatures.](image)

### 5.2 Al-Cu-Mg system

Al-Cu-Mg alloys are important in structural applications where combination of good damage tolerance and high strength is required. In this section, we try to estimate the
transformation kinetics of the $S$ ($\text{Al}_2\text{CuMg}$) phase in Al phase during solution heat treatment. The initial concentrations of Cu and Mg in the Al phase are constant and equal to 0.5 wt% and 1.2 wt%, respectively. The concentrations of Cu and Mg in the S stoichiometric phase are 44.8 wt% and 17.14 wt%, respectively. The position of the initial interface is 6 $\mu$m.

The total number of nodes in FTM is 115, whereas the number of nodes in the S phase and Al phase are 15 and 100, respectively. The thermodynamic condition for the interface concentrations in the Al phase is used from Eq.(16). If we use this interface condition, additional assumptions are not needed. The interface kinetic coefficient divided by entropy difference between phases is $5.0 \times 10^{-14}$ m$^4$/Js. The time-step lengths in the TDA and PFM are $1 \times 10^{-1}$ s and $2 \times 10^{-4}$ s, respectively.

The interface positions during isothermal DC dissolution of the $S$ phase in Al phase at three homogenisation temperatures are presented in Fig.(3). Homogenisation temperatures are 450°C, 470°C and 490°C. The interface concentrations of alloying components in the Al phase during homogenisation computed by the models are presented in Fig.(4). The dissolution kinetics computed by the models are in excellent agreement. The accuracy of the PFM results depends on the quality of the interpolation scheme for computing the driving force.

The numerical results of interface positions and the differences between results in every 360 s are presented in Tab.(1). In every time the difference between the models is extremely low. The order of the differences is $10^{-3}$%. The interface concentrations of Cu
and Mg in the Al phase in every 720 s are written in Tab.(2), only because of the space contribution. The order of the differences of interface concentrations is the same as in the interface position example, $10^{-3}$%. 5 significant numbers of these values are written in tables.

<table>
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<th>interface position (m)</th>
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<tr>
<td>1000</td>
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<td>2000</td>
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<td>3000</td>
<td>5.4E-06</td>
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<td></td>
</tr>
</tbody>
</table>

Figure 3: The interface positions during isothermal DC dissolution of the S phase in Al phase at three homogenisation temperatures.

### 5.3 Al-Mg-Si system

Aluminium alloys of the 6xxx heat-treatable series are the most widely used for the production of extruded parts. Al-Mg-Si system defines the 6xxx series of aluminium alloys. Extrudability of these alloys is strongly influenced by the amount of Mg and Si in solid solution, and the size of Mg$_2$Si phase precipitate particles.

The initial concentrations of Mg and Si in the Al phase are constant and equal to 0.01 wt% and 0.01 wt%, respectively. The concentrations of Mg and Si in the Mg$_2$Si stoichiometric phase are 63.38 wt% and 36.62 wt%, respectively. The position of the initial interface and the number of nodes in the FTM are the same as in the Al-Cu-Mg system. The Vermolen model is used as the reference solution. The model uses the interface condition Eq.(17). The interface kinetic coefficient divided by entropy difference between phases is $5.0 \times 10^{-15}$ m$^4$/Js. The time-step lengths in the TDA and PFM are $1 \times 10^{-1}$ s and $1 \times 10^{-4}$ s, respectively.

The interface positions during isothermal DC dissolution of the Mg$_2$Si phase in Al phase at three homogenisation temperatures are presented in Fig.(5). Homogenisation
Table 1: Numerical data for Fig.(3) and differences between the results of both methods.

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<th>PFM $l$, $\mu$m</th>
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<th>FTM $l$, $\mu$m</th>
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Table 2: Numerical data for Fig.(4) and differences between the results of both methods.

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<th>FTM $C_{\text{Mg}}$, wt.%</th>
<th>PFM $C_{\text{Cu}}$, wt.%</th>
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Figure 4: The interface concentrations of Cu and Mg in the Al phase during isothermal DC dissolution of the S phase in Al phase at three homogenisation temperatures.

<table>
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<td>$\Delta_{\text{max}} \times 10^2, %$</td>
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Table 3: The average and maximum differences between numerical results of the interface concentrations of Mg and Si in the Al phase during isothermal dissolution of the Mg$_2$Si in Al phase at three homogenisation temperatures.

The average ($\Delta_{\text{ave}}$) and maximum differences ($\Delta_{\text{max}}$) are computed from differences in every 360 s. The average differences of the interface position for homogenisation temperatures 520°C, 540°C, 560°C are $2.2680 \times 10^{-3}$%, $2.2652 \times 10^{-3}$%, $3.2909 \times 10^{-3}$%, respectively. The maximum differences ($\Delta_{\text{max}}$) are $4.4394 \times 10^{-3}$%, $5.3058 \times 10^{-3}$% and $5.1368 \times 10^{-3}$%. The average and maximum differences between numerical results of the interface concentrations of Mg and Si in the Al phase are written in Tab.(3).
Figure 5: The interface positions during isothermal DC dissolution of the Mg$_2$Si phase in Al phase at three homogenisation temperatures.

Figure 6: The interface concentrations of Mg and Si in the Al phase during isothermal DC dissolution of the Mg$_2$Si phase in Al phase at three homogenisation temperatures.
6 CONCLUSIONS

The phase transformations in heat treatment processes are modelled by the two-phase approach and phase-field approach. These approaches are connected with the thermodynamic database for aluminium alloys JMatPro. A thermodynamic database is necessary for quantitative simulation of phase transformations. A general two-phase approach for diffusion-controlled phase transformations in multicomponent alloys is proposed. The isothermal diffusion-controlled dissolution of primary nonstoichiometric and stoichiometric particles in aluminium phase is successfully simulated by the two-phase approach and phase-field approach. The whole spectra of represented numerical results, computed by both physical approaches are in excellent agreement. An accurate interpolation or approximation of data from an thermodynamic database is essential for accurate computation of phase transformations by the phase-field approach. The very accurate interpolation by the radial basis functions is used for transformation $\theta$ phase in Al-Cu system. The concentrations in phases at the diffuse interface can be solved in this way. The isothermal diffusion-controlled dissolution of phase S phase in aluminium phase in Al-Cu-Mg system is computed. The very accurate agreement between the results computed by the presented phase-field approach and the Vermolen model for the dissolution of Mg$_2$Si particle in aluminium matrix in Al-Mg-Si system is presented at the end. Our future work is devoted to simulation of the realistic microstructure by the phase-field approach.

REFERENCES


