



Two-dimensional simulation of reactive diffusion in binary systems



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ABSTRACT

Very recently the authors have shown (Svoboda and Fischer, 2013) that the concept of reactive diffusion can successfully be applied to the simulation of one-dimensional diffusive phase transformations in binary systems. The concept is now generalized to two dimensions and used for simulations of diffusive phase transformations in multi-phase binary systems. The kinetics of two systems with different starting configurations and kinetic coefficients is simulated. The simulations show that the concept is robust and the interfaces remain sharp. The comparison with the well-known Cahn–Hilliard phase field method indicates that the present reactive diffusion concept represents a special case of the Cahn–Hilliard phase field method with an infinite value of the interface mobility and zero interface energy. The reactive diffusion concept is, thus, applicable to cases, where the changes of the volume fraction of individual phases dominate their coarsening.

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1. Introduction and motivation

Understanding of the development of several (intermetallic) phases formed in a binary system of elements A and B is mostly based on one-dimensional configurations of a diffusion couple with a crystal rich in element A of one phase in the left part and a crystal rich in element B of another phase in the right part of the specimen. An established concept is that by Dayananda and Murch [1] and later van Loo [2], based on measuring the kinetics of composition profiles, for further details see the recent paper by Paul [3], Section 3 there, and the corresponding references. The main task is the determination of a proper interdiffusion coefficient, see [3]. For sake of completeness and giving some specific applications we mention the following papers: [4] for a Si–Ta system, [5] for a Si–Ti system, [6] for Fe–Nb, [7] and [8] for Ni–Al and Ag–Zn, [9] for Ni–Al, [10], [11] and [12] for a Co–Si system and [13] for Au–Sb system, [14] for Al–Mg and [15] as a review on several systems. It should be mentioned that Paul extended the concept also to quasi-ternary systems, see [3] and applications for a Ni–Co–Pt and Ni–Co–Fe system [16]. An equivalent concept to that of Dayananda/van Loo was presented even half a century ago by Sauer and Freise [17] and confirmed by den Broeder [18], see the very recent paper by Mehrer and Sprengel [19] with applications

to a Co–Nb, Ni–Nb, Al–Mo and Ni–Al systems and the paper by Cui et al. [20] studying a Co–Al system.

With respect to multi-component multi-phase systems and one-dimensional configurations we would like to mention two further approaches:

The first one is the engagement of the DICTRA concept (<http://www.thermocalc.com/DICTRA.htm>), see also [21], which is based on standard linear non-equilibrium thermodynamics assuming local equilibrium at interfaces. As second one we consider the successful application of the Thermodynamic Extremal Principle (TEP), see e.g. [22], for the understanding of the Kirkendall effect in multi-phase systems. We mention here applications for binary systems with multiple stoichiometric intermetallic phases, [23], the extension to multicomponent systems [24], a further extension with modelling the role of sources and sinks for vacancies [25] and the treatment of a system with combined grain boundary and bulk diffusion in the recent paper [26] for Cu–Sn-system. It should be emphasized that the concept based on TEP assumes strictly stoichiometric phases which are separated by sharp phase interfaces. At the interfaces the mass-conservation relation for a moving interface in form of a jump condition, see e.g. [27] and Section 3.2 of [28], must rigorously be fulfilled. The role of sources and sinks for vacancies is manifested in the TEP concept by an additional term α representing the vacancy generation rate density, spanning the whole range from no sources and sinks ($\alpha \equiv 0$) to ideal sources and sinks. We would like to mention that Danielewski, Wierzbna et al. have introduced a somewhat related concept by their

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bi-velocity model, see e.g. the very recent papers [29,30]. However, an explicit involvement of the annihilation/generation of vacancies seems to be the physically proper way.

From our point of view the most successful concept up to now for a one-dimensional setting is the so-called “reactive diffusion concept” (RDC), which is based on a modified form of a proposal by Erdelyi and Schmitz [31], see the previous paper [28], according to which we have received several successful simulations, see again [28].

Let us repeat the advantages of the RDC as

- (i) It works only with thermodynamic quantities (e.g. chemical potentials, diffusion coefficients, diffusive fluxes etc.) without the necessity to handle explicitly the individual phases.
- (ii) It is very simple in treating since the RDC does not make it necessary to engage a jump condition at interfaces between phases, which means that the according algorithm automatically finds the positions of the phase interfaces as steep concentration gradients.

However, the RDC has been formulated and applied up to now only for a one-dimensional configuration. An open research task is now to simulate reactive diffusion processes also in multi-dimensional configurations.

One of the most promising concepts to handle a multi-dimensional configuration is the Phase-Field Method (PFM), see e.g. the overviews by Steinbach [32,33] for applications in materials science and the paper by the co-workers of Steinmann [34] for its formulation in the frame of the finite element method. However, there are also some difficulties in relating the PFM quantities to the thermodynamic ones, see e.g. Wu et al. [35]. A very detailed explanation of the link between PFM and thermodynamics can be taken from the recent paper [36]. Most of the PFM applications are reported for two-dimensional configurations as the recent paper by Cogswell and Carter [37] for ternary systems and four phases, by Kim et al. [38] for binary systems and specific material systems as Ag–Cu, by Böhme et al. [39] and by Wu et al. [35] for a Ni–Al–Pt system. An interesting contribution by Heo et al. [40] can also be mentioned, who incorporated nuclei for interfaces.

For sake of completeness it should be mentioned that also the Monte Carlo method allows a multi-dimensional generalization, see e.g. the recent contribution by Gusak and co-workers [41].

The research goal of this paper is now the multi-dimensional generalization of the very effective RDC, as introduced in a recent paper [28], mainly due to its fascinating simplicity.

2. Equations of reactive diffusion

We consider two components A and B, which may form several intermetallic phases A_yB_{1-y} . Mainly to avoid any deformation state due to the diffusion process we assume that no sources and sinks for vacancies exist in the system and the molar volume Ω is independent of chemical composition and phase. The site fractions of A and B are denoted as y and $1 - y$, resp. To both components the chemical potentials μ_A and μ_B are addressed. Furthermore, the components diffuse with the fluxes \mathbf{j}_A and \mathbf{j}_B through the lattices of individual phases and interfaces. Two conservation equations exist, namely for

$$\text{mass: } \dot{y} = -\Omega \operatorname{div}(\mathbf{j}_A) \quad (1)$$

and

$$\text{lattice positions: } \mathbf{j}_A + \mathbf{j}_B = 0; \quad (2)$$

for details, see, e.g., [42], Section 2 there. A dot above a variable, e.g. \dot{y} , denotes the total derivative with respect to time t . With $\operatorname{grad}(\cdot)$

and $\operatorname{div}(\cdot)$ we denote the gradient and divergence operators applied to a quantity (\cdot) with respect to the actual system. The constitutive equations for the fluxes can be formulated in a common way relating the fluxes with gradients of the chemical potentials. The according derivation can be found in the Appendix and yields

$$\mathbf{j}_A = -\frac{y(1-y)}{RT\Omega} \cdot \frac{D_A D_B}{(yD_A + (1-y)D_B)} \cdot \operatorname{grad}(\mu_A - \mu_B). \quad (3)$$

The gas constant is denoted as R , the absolute temperature as T , and D_A and D_B represent the tracer diffusion coefficients of components A and B in respective phases. The second term on the r.h.s. of Eq. (3) represents a so-called “interdiffusion” coefficient D^{NP} , see the Appendix with respect to its derivation and the superscript “NP”, yielding the diffusion equation in the form

$$RT\Omega \mathbf{j}_A = -y(1-y)D^{\text{NP}} \operatorname{grad}(\mu_A - \mu_B). \quad (4.1)$$

For ideal solutions of components A and B one finds the classical Fick’s first law as

$$\Omega \mathbf{j}_A = -D^{\text{NP}} \operatorname{grad}(y). \quad (4.2)$$

Combination of Eqs. (1) and (4.2) allows formulating the diffusion equation as

$$\dot{y} = \operatorname{div}(D^{\text{NP}} \operatorname{grad}(y)). \quad (4.3)$$

The “interdiffusion” coefficient D^{NP} , however, depends on y , see Eq. (3), and, therefore, a nonlinear partial differential equation must be dealt with. Only for (nearly) identical tracer diffusion coefficients the problem formulation becomes linear in y .

We consider a quadratic region with a periodic boundary condition and a given distribution of the components A (site fraction y) and B (site fraction $1 - y$) in this region as initial condition. The real demanding task, however, is to meet correctly the separation of the total region into sub-regions with different chemical composition and phases. As contact condition at the interface, represented by complicated curves, the equality of chemical potentials must be taken into account, see Eq. (7) below. A detailed description of the substructure and its evolution is given in Section 3.

In the following context we assume as initial phase a solid solution (superscript SS) of components A and B, which allows formulating the chemical potential in the frame of ideal solution as

$$\text{Phase SS: } \mu_A^{\text{SS}} = RT \ln(y), \quad \mu_B^{\text{SS}} = RT \ln(1-y). \quad (5)$$

Moreover, we consider two intermetallic phases A_yB_{1-y} denoted as IMP1 and IMP2. The molar Gibbs energy diagram for all phases SS, IMP1 and IMP2 is depicted in Fig. 1 by using the parameters given by Eq. (11), see later. The chemical potentials are defined according to the concept presented in [28] for both intermetallic phases IMP1 and IMP2 as

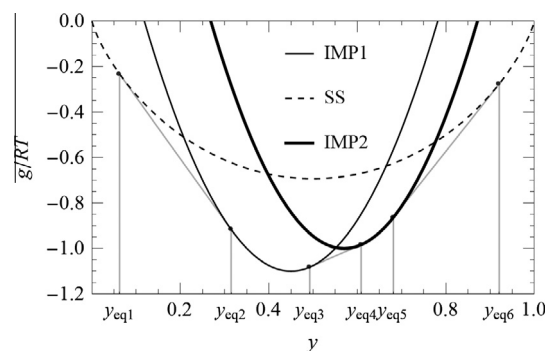


Fig. 1. Molar Gibbs energy diagram for SS, IMP1 and IMP phases plotted for the values of thermodynamic parameters given by Eq. (11). The solubility limits $y_{\text{eq}1}$ to $y_{\text{eq}6}$ are given by the double-tangent construction.

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