Multi-ternary extrapolation scheme for efficient coupling of thermodynamic data to a multi-phase-field model

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ABSTRACT

Phase-field simulation of technical multicomponent and multiphase alloys requires an efficient coupling to thermodynamic databases. Sophisticated interpolation or extrapolation methods allow reducing the frequency of time-intensive thermodynamic (quasi-)equilibrium calculations as has been shown for the multi-binary extrapolation scheme which already has proved to be successful. This paper presents a new multi-ternary extrapolation scheme which better approximates solute redistribution for non-dilute alloys as compared to the multi-binary scheme. Furthermore, numerical problems like multi-binary miscibility gaps can be effectively avoided. The new multi-ternary scheme is derived from the general multicomponent quasi-equilibrium approach and contrasted to the simpler multi-binary redistribution scheme. A quantitative comparison of the two methods is presented for solidification in the ternary system Al–Cr–Ni. The application to a commercial Ni-base superalloy finally demonstrates the potential for simulating multiphase equilibria in composition regions, in which the multi-binary approximation often fails.

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1. Introduction

Phase-field models have gained more and more importance for microstructure modelling in material science. Many different models have been proposed, starting from two-phase models [1–4] up to multiphase-field approaches [5–8] which in addition allow for simulation of the interaction of multiple grains or phases.

The treatment of phase transformation in alloys requires the introduction of concentration fields. Two major approaches can be distinguished: One class of phase-field models for alloys is based on the classical approach for binary alloys of Wheeler et al. [1]. Only a single continuous composition field c is defined, and the free energy density g of the alloy is extrapolated as a mixture in composition of the energy densities of the pure phases. While providing simplicity, this approach has serious disadvantages in applied numerical simulations, like spurious interfacial-chemical contributions which do not allow scaling the interface width independently of other parameters [4,8,9].

A second class of models is based on the multiphase-field approach by Tiaden et al. [8] and includes the multicomponent multiphase-field model described in this paper. This approach separates the total composition c^i in the interface regions into phase compositions c_α^i and thus can also be used for applied simulation on the μm scale. The phase compositions are interrelated by a quasi-equilibrium condition, as indicated earlier by Kim et al. [4]. In contrast to local equilibrium, the quasi-equilibrium approach assumes finite interface mobility and introduces the driving force ΔG as an additional degree of freedom.

While the thermodynamic properties of binary or dilute multicomponent alloys have been successfully described by linear phase diagrams or tabulated tie line data [8,10], high-alloyed materials with strong interactions between the alloying elements require more comprehensive thermodynamic descriptions. Today, databases which have been assessed by the Calphad approach [11] are available for most classes of technical alloys. Consequently, direct coupling of the multicomponent multiphase-field model to Calphad databases [9,12] has been realized in the software MICRESS [13] and has allowed for simulations of phase transformations in technical alloys [14–16].

To keep simulation time in convenient limits, the frequency of accessing the database has to be reduced as much as possible, which can be achieved by using effective interpolation techniques. The improvement of these techniques is subject of the present paper. Three different approaches are distinguished, (a) a "multi-binary" extrapolation scheme which has been developed in [9], (b) a simple "diagonal" approximation, and (c) a new

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"multi-ternary" extrapolation scheme. All three approaches are systematically derived and compared for a ternary Al–Cr–Ni test case with respect to accuracy, numerical stability and computational performance. Finally, at the example of the technical Ni-based superalloy CMSX-4, the advantages and drawbacks for simulation of complex multicomponent systems are discussed.

2. Multiphase-field model with multicomponent solute diffusion

The multiphase-field model used in this work [9] has been developed with the aim to perform simulations of technical solidification and phase transformation processes of metallic alloys. To be suitable for this aim, a formulation has been chosen which splits the chemical composition at interfaces into the individual phase compositions and avoids implicit chemical contributions to the interface energy which scale with the interface thickness. Thus, this formulation is not restricted to the length scale of atomistic diffuse interfaces but allows for application at arbitrary scales.

The possibility of coupling to arbitrary thermodynamic databases is a further characteristic feature of our approach. This is obtained by splitting the free energy density functional into an interfacial term on one hand, and an independent chemical contribution which links to the thermodynamic description on the other hand. Especially the fact that this chemical contribution is formulated as a function of the phase composition c_α and not the mixture composition c (Eq. (2)) turns out to be essential for phases which are stoichiometric or have a restricted solubility range for certain elements.

The multiphase-field model described here is implemented in version 6.154 of the software MICRESS®. All simulations presented in this paper have been performed using this code.

2.1. Model equations

In multiphase-field theory, the parameters $\phi_\alpha(\vec{x}, t)$ describe the spatial distribution of different grains (with different crystallographic orientation) or of different phases with individual thermodynamic properties. At the interfaces, these variables change continuously over an interface thickness η which can be large compared to the atomic interface thickness, but which should always be smaller than the characteristic microstructural features. The local time evolution of $\phi_\alpha(\vec{x}, t)$ is derived from the free energy functional F which integrates the density functional f over the domain Ω [6,9]:

$$F(\{\phi_\alpha\}, \{\vec{c}_\alpha\}) = \int_\Omega f(\{\phi_\alpha\}, \{\vec{c}_\alpha\}) \tag{1}$$

The density functional f can be divided into an interface energy density f^{int} and a chemical free energy contribution f^{chem}. Both are functions of the phase-field variables $\phi_{\alpha=1,\ldots,\nu}(\vec{x}, t)$ of the system with the constraint $\Sigma_\alpha \phi_\alpha = 1$ [6]:

$$
\begin{aligned}
f &= f^{int}(\{\phi_\alpha\}) + f^{chem}(\{\phi_\alpha\}, \{\vec{c}_\alpha\}) \\
&= \sum_{\alpha=1}^{\nu} \sum_{\beta \neq \alpha}^{\nu} \frac{4\sigma_{\alpha\beta}}{\nu\eta} \left(-\frac{\eta^2}{\pi^2} \nabla\phi_\alpha \nabla\phi_\beta + \phi_\alpha \phi_\beta \right) + \sum_{\alpha=1}^{\nu} \phi_\alpha f_\alpha(\vec{c}_\alpha)
\end{aligned}
\tag{2}
$$

Here, $\sigma_{\alpha\beta}$ denotes the anisotropic energy of the interface between α and β, η is the thickness of the phase boundary which is identical for all interfaces, and ν is the number of locally coexisting phases [6]. The multiphase-field equation, which describes the time evolution of $\phi_\alpha(\vec{x}, t)$ in multiple phase transformations, is derived by minimization of the total free energy F following the relaxation principle

$$\dot{\phi}_\alpha = -\sum_{\beta=1}^{\nu} \widetilde{M}_{\alpha\beta}(\vec{n}) \left(\frac{\delta F}{\delta \phi_\alpha} - \frac{\delta F}{\delta \phi_\beta} \right) \tag{3}$$

where $\widetilde{M}_{\alpha\beta}(\vec{n})$ are anisotropic kinetic constants. Finally, the general form of the multiphase-field equations is obtained [9]:

$$
\begin{aligned}
\dot{\phi}_\alpha = \sum_{\beta \neq \alpha}^{\nu} M_{\alpha\beta} &\left[\frac{\sigma_{\alpha\beta}}{\nu} \left(\frac{1}{2} (\nabla^2\phi_\alpha - \nabla^2\phi_\beta) + \frac{\pi^2}{2\eta_{\alpha\beta}^2} (\phi_\beta - \phi_\alpha) \right) \right. \\
&\left. + \sum_{\alpha \neq \beta \neq \gamma}^{n} J_{\alpha\beta\gamma} + |\nabla\phi| \Delta G_{\alpha\beta} \right]
\end{aligned}
\tag{4}
$$

with:

$$J_{\alpha\beta\gamma} = \frac{1}{2}(\sigma_{\beta\gamma} - \sigma_{\alpha\gamma})\left(\frac{\pi^2}{\eta^2}\phi_\gamma + \nabla^2\phi_\gamma \right)$$

$$M_{\alpha\beta}(\vec{n}) = \widetilde{M}_{\alpha\beta}(\vec{n}) \frac{8\eta}{\pi^2}$$

For sake of simplicity, an isotropic formulation is shown here. The corresponding equations including anisotropy are given in [17]. $\Delta G_{\alpha\beta}$ is the thermodynamic driving force which is a function of temperature T and local composition $\vec{c}$ and which has been scaled by an interpolation function to locally recover the Gibbs–Thompson relation. From Eqs. (2) and (3) $\Delta G_{\alpha\beta}$ can be generally expressed as

$$\Delta G_{\alpha\beta} = g_\beta - g_\alpha - \sum_k \tilde{\mu}^k(c_\beta^k - c_\alpha^k) \tag{5}$$

However, the Lagrange multipliers $\tilde{\mu}^k$ cannot be determined without further assumptions. Their physical meaning will be specified in Section 2.2. In Eq. (5) as well as in the following, molar free energy densities f_α have been replaced by molar Gibbs energy densities g_α. This is allowed in case of constant molar volumes, which is a typical assumption in many phase-field models. We prefer using Gibbs energy densities for the chemical part of the functional because Calphad databases are formulated as function of pressure and temperature. $\Delta G_{\alpha\beta}$ which is the only concentration dependent quantity of the multiphase-field equation couples the phase-field equations to the multiphase diffusion equations for n elements:

$$\dot{c}^k = \nabla \sum_{\alpha=1}^{\nu} \sum_{l=1}^{n-1} \phi_\alpha D_\alpha^{kl} \nabla c_\alpha^l \quad \text{with} \quad D_\alpha^{kl} = D_\alpha^{kl,0} \exp\left(\frac{Q_\alpha^{kl}}{RT} \right) \tag{6}$$

This equation is solved for $n-1$ dissolved elements by an explicit scheme. $D_\alpha^{kl,0}$ and Q_α^{kl} are calculated online using a mobility database for the given concentration and temperature. How the phase-field increments $\dot{\phi}_\alpha$ couple back to Eq. (6) is discussed in the following.

2.2. Quasi-equilibrium approach

In almost all solidification processes, but also in many solid–solid phase transformations, the conditions at the phase boundaries are relatively close to local thermodynamic equilibrium, while the global state is clearly off-equilibrium. This is reflected in many popular micro-segregation models like e.g. Scheil or Clyne-Kurz [18]. Also the software Dictra [19] uses local equilibrium as condition at interfaces. Nevertheless, most phase-field models allow for small deviations from equilibrium. This seems appropriate not only as it provides the driving force ΔG for the phase-field Eq. (4): It also allows for finite interface kinetics and for curvature undercooling, and, maybe most important, for reduction of computational effort by providing an explicit numerical scheme.