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Application of the thermodynamic extremal principle to phase-field modeling of non-equilibrium solidification in multi-component alloys



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ABSTRACT

Modeling of non-equilibrium solidification in multi-component alloys is of singular importance in microstructure control, which however owing to the complex systems with complex additional constraints is still an open problem. In this work, the thermodynamic extremal principle was applied to solve the complex additional constraints self-consistently in thermodynamics. Consequently, short-range solute redistribution and long-range solute diffusion that share the same mobility are integrated naturally into the solute diffusion equations, thus avoiding the introduction of additional kinetic coefficients (e.g. interface permeability) to describe solute redistribution. Application to the non-equilibrium solidification of Al-Si-Cu alloys shows that anomalous solute trapping and anomalous solute profiles within the diffuse interface could occur, thus highlighting the important effect of the interaction among the component elements on the interface kinetics. The current phase-field model might be preferred for simulations not only because of its simplest form of evolution equations but also its feasibility to increase the simulation efficiency by the “thin interface limit” analysis.

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1. Introduction

Most commercial materials are multi-component alloys (e.g. the Ni-based superalloy may consist of ten elements such as Ni, Al, Co, Cr, Mo, Ta, W, Re, Ru and Hf [1]), whose properties are determined by the microstructure that develops during solidification and subsequent processing routes [2]. This is also the case for high-entropy alloys (HEAs) that are based on the multiple principal elements rather than just one or two in the conventional alloys [3–5]. With the increase of the number of components, the variables in the modeling system increase and so are the additional constraints among them. The interaction among the component elements could be so strong that the alloys cannot be approximated as dilute solutions anymore and the kinetic processes could be so sluggish (e.g. the sluggish diffusion effect in HEAs [6,7]) that deviation from local-equilibrium condition is inevitable at the migrating interface. Modeling of non-equilibrium solidification in multi-component alloys therefore is of singular importance in microstructure control, which however owing to the complex systems with complex additional constraints is still an open problem.

By introducing a phase-field (ϕ) that is constant (e.g. 0 or 1) in the bulk phases and changes continuously (e.g. from 0 to 1) across the interface, phase-field modeling avoids the complex front

tracking procedure in the sharp interface models and become a powerful simulation tool for the prediction of microstructure formation [8–10]. For solidification of multi-component alloys, there are two kinds of additional constraints in the modeling system. One is the single-phase state at any point in the bulk phases:

$$\phi_S + \phi_L = 1, \quad (1)$$

where ϕ_i ($i = S, L$) is the phase-field. The other is the mixture law for the solute molar fractions:

$$c^j = h_S c_S^j + h_L c_L^j \quad (j = 1, 2, \dots, n), \quad (2)$$

where h_i ($h_S + h_L = 1$) is the interpolation function, c^j , c_S^j and c_L^j are the overall, solid and liquid solute molar fractions of component j . One can see that for the system of $n + 1$ components where $j = 0$ and $j = 1, 2, \dots, n$ denote the solvent and the solutes, there are totally $n + 1$ additional constraints.

Because the classical Cahn-Hilliard [11] and Allen-Cahn [12] equations are able to deal with the first but not the second kind of additional constraints [13], several approximate treatments were introduced to propose the phase-field models (PFMs). Initially, a third kind of additional constraints was introduced to obtain the evolution equations of phase-field (ϕ) and overall solute molar fractions (c^j). One treatment is the condition of equal solute molar

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fractions [14]. The interface is a mixture of solid and liquid with the same solute molar fractions ($c_S^j = c_L^j = c^j$) but different diffusion potentials ($\tilde{\mu}_S^j \neq \tilde{\mu}_L^j$) [15]. The bulk and interface contributions however cannot be decoupled under equilibrium conditions and it is not that convenient to improve the simulation efficiency by the “thin interface limit” analysis [16]. The other treatment is the condition of equal diffusion potentials [17]. The interface is a mixture of solid and liquid with different solute molar fractions ($c_S^j \neq c_L^j$) but the same diffusion potentials ($\tilde{\mu}_S^j = \tilde{\mu}_L^j$) [18,19]. Even though the bulk and interface contributions can be decoupled under equilibrium conditions, n additional non-linear equations (i.e. $\tilde{\mu}_S^j = \tilde{\mu}_L^j$ ($j = 1, 2, \dots, n$)) need to be solved at each point within the diffuse interface, thus resulting in significant simulation complexity especially for the concentrated alloys.

Steinbach et al. [20] proposed recently a novel concept of finite interface dissipation. If solute diffusion in solid and liquid is split into the long-range and short-range fluctuations with a specific cut-off wave vector by the Fourier transform, a sum of all the short-wavelength fluctuations is the flux of solute redistribution between solid and liquid [9,21]. Its dissipation was assumed to be controlled by interface permeability p , e.g. $p \rightarrow \infty$ corresponds to an infinite fast solute redistribution [9,20,21] (i.e. the condition of equal diffusion potentials). Such PFM with finite interface dissipation was extended subsequently to multi-component multi-phase alloys [22,23]. In this case, the interface is a mixture of solid and liquid with different solute molar fractions ($c_S^j \neq c_L^j$) and different diffusion potentials ($\tilde{\mu}_S^j \neq \tilde{\mu}_L^j$). The evolution equations are derived for the phase-fields, the solid and liquid solute molar fractions, thus avoiding the significant simulation complexity due to the introduction of the condition of equal diffusion potentials. It should be pointed out however that Steinbach et al. [20,22] tried to solve the additional constraints Eq. (2) by the Lagrange multiplier method. After dividing the system into distinct reference volumes (RVs), they assumed firstly that each RV is isolated to formulate short-range solute redistribution by the Allen-Cahn equation [12] and the overall solute concentration is time independent to obtain the Lagrange multiplier. After that, long-range solute diffusion between the adjacent RVs is added to obtain the solute diffusion equations. Such recipe as summarized in Fig. 1a can only solve the additional constraints Eq. (2) approximatively, which is the reason why additionally kinetic coefficients (e.g. interface permeability) should be introduced.

Recently, the thermodynamic extremal principle (TEP) [24,25] has been developed to be a handy tool for modeling of complex thermodynamic systems with complex additional constraints. So far, it has been applied successfully to diffusion and creep [26], precipitation [27], diffusion-controlled phase-transformation [28,29], grain growth and coarsening [30], rapid solidification [31,32] etc. Applications of TEP [24,25] to phase-field modeling show that the standard phase-field equations can be reproduced [33] and both the first and second kinds of additional constraints can be solved self-consistently in thermodynamics for binary alloys [13,34]. Short-range solute redistribution as well as its finite dissipation is inherent in such PFMs [13,34] and for any infinitesimal volume chosen within the interface, there is a “sharp interface” between solid and liquid at which solute and chemical potential jump happens. Furthermore, interface permeability is not introduced and the three evolution equations for the phase-field, solute molar fractions in solid and liquid follow the Onsager's reciprocal relation. Since TEP [24,25] is able to solve the complex additional constraints and derive the evolution equations in a strict mathematical and physical way, it should be the recipe for phase-field modeling of non-equilibrium solidification in multi-

component alloys.

In this work, TEP [24,25] is applied to phase-field modeling of non-equilibrium solidification in multi-component alloys. First, the modeling system is described and two sets of evolution equations are formulated, i.e. one in the form of phenomenological equations [35] in which the fluxes and their conjugated driving forces are linearly related and the other not (Section 2). The former is clear in physical meaning but complex in form, whereas, the latter is much simpler than all the previous PFMs for multi-component alloys [15,18,19,22]. After that, the interface dissipations in the current PFM are discussed and the interface conditions are analyzed in the case of 1-D steady-state growth (Section 3). It is shown that the adoption of effective mobilities for non-equilibrium solute diffusion [34] allows not only long-range solute diffusion but also short-range solute redistribution under non-equilibrium conditions. Application to the non-equilibrium solidification of Al-Si-Cu ternary alloy (Section 4.1) shows that anomalous solute trapping and solute profiles within the diffuse interface happen if the interaction among the component elements is strong enough. Through a comparison study with the previous work on both sharp-interface modeling and phase-field modeling (Section 4.2), the validity and limitation of current model are discussed. Finally, the current work is summarized in Section 5.

2. The model

2.1. Problem descriptions

The modeling system with a volume Ω consists of two phases solid and liquid and $n + 1$ components. For simplicity, the molar volume of each component in solid and liquid is assumed to be the same v_m . In the current work, it is the multi-component alloys but not the multi-component multi-phase alloys that is chosen because it is the second but not the first kind of additional constraints that cannot be solved self-consistently in thermodynamics. Furthermore, it is the design of HEAs with a single solid-solution phase [36–38] but not multi-phases that is of more extensive concern for the development of high performance HEAs. The total Gibbs free energy of the system is the integral of free energy density over the system volume Ω , i.e.

$$\tilde{F} = F + F_{ad} = \int_{\Omega} (f + f_{ad}) d\Omega = \int_{\Omega} (f_{intf} + f_{bulk} + f_{ad}) d\Omega, \quad (3)$$

where the contributions from the interface f_{intf} , bulk phases f_{bulk} and additional constraint f_{ad} due to Eq. (2) ¹ are:

$$f_{intf} = \frac{4\sigma_{SL}}{\eta} \left[\frac{\eta^2}{\pi^2} (\nabla\phi)^2 + \phi(1-\phi) \right], \quad (4)$$

$$f_{bulk} = \frac{1}{v_m} \sum_{i=S,L} h_i g_i = \frac{1}{v_m} \sum_{i=S,L} h_i \sum_{j=0}^n \mu_i^j c_i^j, \quad (5)$$

$$f_{ad} = \sum_{j=1}^n \lambda^j \left[c^j - (h_S c_S^j + h_L c_L^j) \right], \quad (6)$$

Here σ_{SL} is the interface energy, f_{intf} is defined by the double-obstacle potential [8] such that the interfacial width η remains

¹ The first kind of additional constraint Eq. (1) has been already incorporated into Eqs. (4)–(6), and thus only the second kind of additional constraints Eq. (2) needs to be considered.

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