

# Morphological stability analysis for planar interface during rapidly directional solidification of concentrated multi-component alloys

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## Abstract

Coupling with the thermodynamic database and incorporating the effects of local non-equilibrium and off-diagonal diffusional interactions, an extended morphological stability analysis was performed for a planar interface upon rapid solidification of concentrated multi-component alloys. Compared with the previous work, the model is able to clarify the effects of concentration dependence and diffusional interactions on the interface stability. Taking the Al–Mg–Zn alloy as an example, the neutral stability, i.e. the critical Zn concentration (with Mg concentration fixed) for the breakdown of the planar interface, was studied. The stability mechanisms for the neutral Zn concentration subjected to different Mg concentrations and the off-diagonal diffusion effect were clarified. In particular, for high Mg concentrations, a stage of absolute instability dependent on the Mg diffusion effect but independent of the Zn concentration happens. Although the off-diagonal diffusion is several orders of magnitude smaller than the solute diffusion coefficients, it can significantly change the Zn concentration for the neutral stability conditions.

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**Keywords:** Interface stability; Multi-component; Concentrated; Rapid solidification

## 1. Introduction

Directional solidification has been proved to be a very powerful method of producing traditional as well as new materials [1,2]. Therefore, its morphological stability has been an important subject for several decades for both scientific and engineering reasons [1,3]. In order to understand the interface stability during directional solidification, a great deal of theoretical work has been carried out, including marginal stability analysis [4,5], microscopic solvability theory [6,7] and phase-field simulation [8]. Among these approaches, the marginal stability theory is widely accepted because of its successful application to dendrite growth [9,10].

Linear stability analysis of a planar interface was initiated by Mullins and Sekerka (MS) [4] for the near-equilibrium solidification of a dilute binary alloy with small Peclet numbers. Dedicated to removing this limitation, Trivedi and Kurz (TK) [5] extended the MS theory to cases of high Peclet number. However, both the MS and TK theories assume local equilibrium diffusion (LED) at the solid/liquid (S/L) interface and are not applicable to rapid solidification where both the S/L interface and the bulk liquid are under local non-equilibrium diffusion (LNED) conditions. Incorporating non-equilibrium interface conditions, a thermodynamically consistent interface kinetic model (IKM) was, according to the solute trapping model of Aziz [11] and Jackson et al. [12], derived by Boettinger et al. [13]. On this basis, a linear stability model was proposed by Huntley and Davis [14], and a reasonable mechanism for steady-state cellular stability was proposed.

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In the above work [4,5,14], LNEED conditions are considered only at the interface, whereas the effect of LNEED in the bulk liquid is not considered, i.e. the diffusion speed is infinite according to the classical diffusion theory. Following extended irreversible thermodynamics (EIT) [15–18], the diffusion speed should be finite to satisfy the LNEED condition in the strongly non-equilibrium system. For example, during rapid solidification, the interface velocity ( $V$ ) can be comparable to or even larger than the diffusion speed of the liquid ( $V_D^L$ ), thus leading to complete solute trapping at a finite  $V = V_D^L$  [16–18]. From EIT, Lee et al. [19] extended the model of Huntley and Davis [14] to the cases of LNEED in the liquid. The model, however, adopts the kinetic liquidus under LED conditions [19] and is thus not self-consistent in thermodynamics. Departing from LNEED theories, Galenko and Danilov [20] carried out a self-consistent analysis for the stability of the planar interface, which yields quantitative agreement with the experimental results of non-equilibrium solidification.

For simplicity, linear liquidus and solidus (LLS) is assumed in the above stability analysis [4,5,14,19,20], which is restricted, however, to very limited alloy systems. For most alloys with typically curved solidus and liquidus, the above models with LLS result in significant deviations. This was pointed out by Divenuti and Ando (DA) [21], who incorporated the effect of non-linear liquidus/solidus (NLLS) into the TK model. Combining the effects of NLLS and LNEED, Wang et al. developed an extended stability model [22] and studied neutral stability conditions [23], subsequently followed by the dendrite growth model [22,24], the dendrite fragmentation model [25] and overall solidification kinetics [26,27]. An extension of the model of Wang et al. [22–24] to concentrated binary alloys was performed recently by combining with a thermodynamic database [28,29].

All the above analysis focuses on binary alloys, while most industrial alloys are concentrated multi-component alloys. It thus becomes inevitable to model morphological stability during solidification of concentrated multi-component alloys. Assuming LED and LLS, diffusional interactions were introduced into the diffusion matrix by Hunziker [30] and Hoyt [31], and a linear stability analysis was carried out. Ludwig et al. [32,33] proposed a general concept for the interface stability of multi-component alloys, but this assumes LED in the bulk liquid and ignores diffusional interactions, and thus is not applicable to the strongly non-equilibrium solidification of concentrated alloys.

Compared with the binary or dilute multi-component alloys, the assumptions of LED and LLS should be relaxed, and the effect due to diffusional interactions should be considered for rapid solidification of concentrated multi-component alloys. On this basis, theoretical modeling for morphological stability during rapid solidification of concentrated multi-component alloys needs to incorporate the effects of LNEED at the interface and in the bulk liquid, thermodynamic database, and off-diagonal terms in the diffusion matrix (i.e. diffusional interactions). Such

a model can demonstrate the effects of concentration dependence and off-diagonal diffusion terms on morphological stability, which still remains an open question [34]. According to Rougier et al. [35], diffusional interactions can significantly affect the precipitation kinetics of  $\gamma'$  in Ni-based superalloy and are very important in the modeling of concentrated multi-component alloys. In the present work, an extended morphological stability model for the planar interface is established, and neutral stability conditions subjected to different effects are analyzed for the Al–Mg–Zn alloy.

## 2. Model

To study the stability of the planar interface, the governing equations at the interface and in the bulk phases, i.e. IKM and the diffusion equation, should be deduced first. Then, subjected to an infinitesimal perturbation, deviations of the physical quantities at the curved S/L interface are obtained through linear corrections. On this basis, the stability criterion (SC) for the breakdown of the planar interface is obtained.

### 2.1. Governing equations and boundary conditions

Following EIT [15], a generalization of the classical Fick's law for mass transport, including the relaxation to LED, is given by

$$J_j + A_j \nabla \mu_j + \tau_j \frac{\partial J_j}{\partial t} = 0 \quad (1)$$

where  $J_j$  is the diffusion flux,  $\tau_j$  the relaxation time of  $J_j$  to its steady state value,  $A_j$  the mobility of solute  $j$ , and  $\mu_j$  the chemical potential of  $j$ .  $A_j$  and  $\tau_j$  are given by

$$A_j = \frac{D_j}{V_m} \left( \frac{\partial \mu_j}{\partial x_j} \right)^{-1} \quad \tau_j = D_j / V_{Dj}^2 \quad (2)$$

where  $D_j$  is the diffusion coefficient of  $j$ ,  $V_{Dj}$  is the diffusion speed of solute  $j$ , and  $V_m$  is the molar volume (assumed to be the same for all species). In the concentrated alloys, the diffusional interactions have to be considered, and Eq. (1) is generalized as

$$J_j + \sum_{k=1}^n \left( A_{jk} \nabla \mu_k + \tau_{jk} \frac{\partial J_k}{\partial t} \right) = 0 \quad (3)$$

with  $A_{jk}$  and  $\tau_{jk}$  as

$$A_{jk} = \frac{D_{jk}}{V_m} \left( \frac{\partial \mu_k}{\partial x_k} \right)^{-1} \quad \tau_{jk} = D_{jk} / V_{Dj}^2 \quad (4)$$

where  $\mathbf{D}$  is the diffusion matrix with its element  $D_{jk}$  representing the interactions between components  $j$  and  $k$ . And  $\tau_{jk}$  is the influence of species  $k$  on the relaxation time of component  $j$ . Then a combination of Eq. (3) with the local mass conservation law

$$\frac{\partial x_j}{\partial t} = -V_m \nabla J_j \quad (5)$$

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