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Phase field and analytical study of mushy zone solidification in a static thermal gradient: From dendrites to planar front

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ABSTRACT

We present phase field simulation results of the directional solidification of an Al–Cu4wt% alloy followed by a holding stage during which the mushy zone solidifies in a static thermal gradient. During the holding stage, the initially dendritic morphology solidifies through the TGZM (Temperature Gradient Zone Melting) process, in accordance with recent in-situ X-ray measurements. This yields a planar solid/liquid interface that evolves towards a partial equilibrium where the liquid is homogeneous and the solid still shows a microsegregation pattern. A slow solid state diffusion then drives the subsequent evolution towards the final stationary state where chemical fluxes vanish. We study analytically the microsegregation in the solid, showing that TGZM has a major influence on its development during directional dendritic growth and on its evolution during the holding stage. Especially, we develop an expression for the concentration profile within a secondary dendrite arm during growth. We also develop theoretical arguments to determine the position in the thermal gradient of a transition region that separates the solid at partial equilibrium into two spatial domains. Close to the solid/liquid interface, the microsegregation exhibits a simple pattern resulting from a full remelting of the dendritic solid, while at lower temperatures the pattern is more complex and results from an only partial remelting.

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1. Introduction

Alloy solidification usually proceeds in a dendritic growth morphology leaving a solid-liquid two-phase mixture called the mushy zone behind the growth front [1]. In isothermal conditions and for alloy concentrations lying in the two-phase region, the phase transformation is completed through an Ostwald ripening regime during which the reduction of the total interface energy within the mushy zone is the main driving force for diffusional processes. Then solid regions presenting small curvature grow at the expense of the ones presenting large curvature, so that a spatially homogeneous distribution of large solid particles is the last stage prior to thermodynamic equilibrium. When the system solidifies in a unidirectional thermal gradient, the situation is different because the isotropy of the space is broken by the unidirectionality of the temperature field. As time goes to infinity, chemical fluxes vanish (while of course heat fluxes do not vanish) and an isothermal solid/liquid interface (i.e. perpendicular to the thermal gradient) establishes, separating the liquid at high temperature from the solid at lower temperature. While the route to equilibrium in isothermal conditions via ripening has been studied since a long time [2], the evolution in a unidirectional thermal gradient has recently received an increasing attention [3–10] promoted by the development of the in-situ synchrotron X-ray radiography technique that allows one to observe the real-time dynamics of the system [11,12]. The understanding of the equilibration path in a thermal gradient is not only an interesting topic on the fundamental level but has also important practical purposes. Indeed, static thermal gradients may exist in situations as diverse as in a Bridgmann furnace, across the sea ice, in a lava lake [13] or at the surface of an exoplanet [14].

One way to study the equilibration in a unidirectional thermal gradient consists in the following two-stages procedure. First, the alloy is directionally solidified during cooling in the thermal gradient leading to a dendritic microstructure. Second, the cooling of the sample is stopped but the thermal gradient is kept (holding stage). Then the mushy zone inherited from the cooling stage solidifies. Together with liquid-film-migration [15], coarsening or thermo-migration [16], the Temperature Gradient Zone Melting (TGZM) process has been recognized to be of primary importance for the mushy zone solidification. Indeed, it allows the liquid

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domains within the mushy zone to migrate towards and finally reach the bulk liquid via solute diffusion in the liquid. This continuous melting/resolidification process provides a very efficient mechanism for the increase of the solid fraction in the mushy zone. However, TGZM does not lead to the final stationary state without chemical flux. As will be shown in this article, a partial equilibrium with a microsegregation pattern in the solid establishes instead, with the route to the final stationary state being ensured by the slower solid state diffusion.

TGZM was first studied theoretically by Pfann [17] and Tiller [18]. It was thought as a powerful process to produce semiconductor devices and single crystals, or to provide a chemical purification of a solidified alloy. TGZM was then recognized to occur during directional solidification, especially yielding a migration of the secondary dendrite arms. Theoretical models were established [19,20] and their experimental validations using in-situ X-ray measurements were undertaken later on [4,21]. TGZM, as a key process for the morphological evolution of a mushy zone, is thus of primary importance for technological casting and should for example influence the so-called melt feeding of the mushy zone. Not only responsible for the migration of secondary dendrite arms, TGZM has also a large influence on the microsegregation pattern in the solid for directionally solidified metals. Thus, it needs further attention in order to understand and predict microsegregation by numerical models and simulations. For example, TGZM would be completely neglected when the directional growth is simulated in a two-dimensional cutting plane perpendicular to the thermal gradient. This is an approximation that is widely used for example for the simulation of microsegregation pattern in directionally solidified Ni-based alloys for turbine blade applications [22]. We will show that the microsegregation within a secondary dendrite arm changes considerably when the arm migrates within the mushy zone due to TGZM.

As mentioned above, TGZM has been recognized to be the main dynamical process when the sample is kept in a static and unidirectional thermal gradient after directional solidification [9]. In this article, we aim at investigating this situation numerically. We use the phase field method to simulate the dendritic growth directional solidification and a subsequent holding stage. These simulations describe the evolution of the solid/liquid interfaces as well as the evolution of the solute concentration field. Not only comparing our results with the quantitative investigations in Ref. [9], we go further with an analysis of the microsegregation pattern in the solid phase.

The phase field method has emerged as the tool of choice to tackle free boundary problems such as the solid-liquid phase transformations we are interested in here. The simulation of directionally grown dendritic solidification pattern has already a long track record, see for example Refs. [23–28], and can be performed today for large three-dimensional arrays [29]. However, only very recently this method was used to study the TGZM process. In Ref. [30], an analytic solution for the motion of a liquid droplet in a static thermal gradient is confronted to phase field simulations, including a situation relatively close to the one that is encountered after directional dendritic solidification as it is of interest in the present paper. However, in Ref. [30], the calculation domain covers only a dozen of secondary arms and does not contain the dendrite tip and the final flat front equilibrium position. Here, we simulate a longer and wider system, allowing us to study the complete scenario of mushy zone solidification.

The paper is organized as follows. In Section 2, we present the experimental observations of Ref. [9] and their interpretation in terms of the TGZM process, whose simplest theoretical form is recalled. In Section 3, we present our simulation of the directional solidification stage for a Al–Cu4wt% alloy. We use theoretical arguments to describe the system at steady state that represents the initial condition for the holding stage. In Section 4, we present the simulation of the holding stage. We interpret our results, compare them with experiment and then interpret the microsegregation pattern in the solid phase. We finally conclude in Section 5.

2. Experimental observations and TGZM process

In Ref. [9], the solidification of the mushy zone in a unidirectional and static thermal gradient (the holding stage) is studied experimentally. Initially the microstructure consists of a solid crust, a region at low temperatures that has totally solidified during the cooling stage, the mushy zone at intermediate temperatures, where the solid phase and the liquid phase coexist, and the bulk liquid at higher temperatures. During the holding stage, the solid crust/mushy zone interface moves upwards to higher temperatures at the expense of the mushy zone. On the other hand, the mushy zone/bulk liquid interface retreats, and after some time, the solid crust/mushy zone and the mushy zone/liquid interfaces merge, which marks a flat front equilibrium state except for a microsegregation still present in the solid. Using X-ray in-situ radiography, the authors investigate quantitatively several observables such as the position of each interface, the concentration on the liquid side of the mushy zone/liquid interface or the concentration profile in the bulk liquid.

In addition to the X-ray measurements, movies of the evolution of the microstructure are also provided (Supplemental material of Ref. [9]). The gray level quantifies the local composition of the sample with a spatial resolution of about 5 μm. While in the solid crust and in the bulk liquid, the variations of the gray level are weak, the mushy zone presents pronounced inhomogeneities allowing to discover the solid-liquid microstructure. These movies reveal that the solidification of the mushy zone mainly takes place through an apparent migration of the whole microstructure upwards in the temperature field. This observation evidences the crucial role of the TGZM process in the solidification of the mushy zone.

The TGZM process is a particularity of the relaxation towards the two-phase equilibrium in a uni-directional thermal gradient and was extensively studied for describing the migration of liquid droplets. Since the equilibrium concentrations vary in a thermal gradient, concentration gradients are present in the liquid phase even with local equilibrium conditions at the solid/liquid interfaces. The corresponding fluxes yield a movement of these interfaces through the Stefan condition:

$$\left(C_L^i - C_S^i\right)V_n = -D(\nabla C_L \cdot \mathbf{n}) \quad (1)$$

where V_n is the normal velocity of the interface, C_L^i and C_S^i are respectively the concentration on the liquid and on the solid side of the interface, D is the diffusion coefficient in the liquid, ∇C_L is the gradient at the interface of the concentration C_L in the liquid and $\mathbf{n}$ is the unit vector normal to the interface. In the case of a linear phase diagram, and neglecting the Gibbs-Thomson effect, the concentration in the liquid phase is equal to the liquidus concentration. The concentration field is thus unidirectional and linearly depending on z, i.e. $C_L = C_L^0(z) = G(z_M - z)/m$ where G is the unidirectional thermal gradient in the z direction (unit vector $\mathbf{z}$), $m > 0$ is the liquidus slope and z_M is the z-position of the melting temperature T_M of the pure material (here pure aluminium). Let us note that even when C_S^i differs from its equilibrium value, the local equilibrium condition at the interface may also be assumed. Indeed, a steep concentration gradient in the solid phase actually allows the interface to be at equilibrium [31]. If now the concentration on the

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