Large scale phase-field simulations of directional ternary eutectic solidification

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Abstract—Pattern formation in ternary eutectic microstructures is difficult to predict because of the complex interactions between diffusion and surface energies, as well as the large number of possible configurations which three phases can assume. The silver–aluminum–copper (Ag–Al–Cu) ternary eutectic is of particular interest to researchers, but its study is complicated by an unusually large solubility change. Due to this change, it is difficult to observe the patterns that form during the solid–liquid phase transition experimentally. This causes significant differences between the structure that forms during solidification and what is observed after traditional directional solidification processing. In order to model the solidification behavior and pattern formation in representative volume elements, large scale phase-field simulations are employed. Two different parameter sets are used. The first set uses the phase fractions and compositions of the as-solidified structure. The other used phase fractions found at lower temperatures in order to be comparable to experimental microstructures available in the literature. The second set of simulation results is compared quantitatively to experimental micrographs and found to be in good agreement. The predicted three-dimensional microstructures for the as-solidified structure are presented.
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1. Introduction

The manipulation of metal alloys to change the properties of the resulting material is a technique that has been practiced for thousands of years. This article helps to improve our understanding of the underlying processes, in order to control the behavior and properties of alloys at the levels required for modern engineering applications. In industrial production, metal casting remains one of the primary means of production, where the properties of the material are in large part determined by the microstructure that forms while the alloy is solidifying. Advances in fields like chemistry, materials science, and computer science, now allow for accurate modeling of increasingly complex solidification behavior. The composition of the alloys and the process conditions themselves have a significant influence on the developing microstructures, whose patterns in turn affect the material properties. Alloys which exhibit a ternary eutectic point are one particular class of alloys in which there is growing interest, in part because their adjustable properties show promise in a variety of industrial applications. They are currently used in a number of lead-free solders [1]. From a scientific point of view they are of interest due to the complex interplay between diffusion, undercooling and surface energies at the solid/liquid interface, they produce a wide variety of microstructural patterns. The observed patterns can be classified by five theoretical patterns that were predicted by Ruggiero et al. [2] based on geometrical considerations for ternary systems, which were later depicted graphically by Lewis et al. [3].

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The focus of the work is the pattern formation during the solidification of the system silver–aluminum–copper (Ag–Al–Cu) around the ternary eutectic point. The system is favored due to a relatively low eutectic temperature (773.6 K) and a roughly equal amount of each solid phase. First experimental observations of ternary eutectic systems, including Ag–Al–Cu, were conducted by Cooksey and Hellawell [4] in 1967. It should be considered that the undercooling velocity was orders of magnitudes larger compared to current experimental studies and the composition of the samples is off-eutectic. The influence of different velocities and compositions was systematically studied by McCartney et al. [5]. Analysis and modeling of the Ag–Al–Cu system is also supported by a detailed thermodynamic description developed by Hecht and Witusiewicz et al. [6–8]. The liquidus projection and the ternary eutectic point E_{cal}, shown in Fig. 1, are based on these thermodynamic results.

One difficulty in analyzing structures in the Ag–Al–Cu system is a result of the unusually large change in solubility of Ag in the Al-phase. The solubility falls by more than 50% (from 16.9% to around 8%) over the 20 K directly below the eutectic temperature [9]. This leads to significant solid state evolution of the microstructure, particularly in directional solidification experiments at the low velocities necessary to maintain coupled growth and a planar interface. As a result, both phase fractions and composition of the Al-phase in directionally solidified samples deviate significantly from equilibrium eutectic values [9–11]. However, experimental results do show that the ratio of experimentally measured phase fractions to each other always corresponds to the equilibrium fractions predicted by the phase diagram in a small range below the eutectic temperature. The original solidification microstructure can only be obtained via quenching experiments, and observation of the sample directly behind the quenched interface. The authors are not aware of such microstructural images currently available in the literature.

The goal of this work is to investigate the pattern formation during solidification of (Ag–Al–Cu) ternary eutectics using phase field modeling. For this we simulate three-dimensional microstructures for comparison with experimental micrographs and analyze the influence of different parameters. In last years, the phase-field method has been established as a powerful tool to simulate free boundary problems, such as solidification [12,13]. The coupling to different physical phenomena enables a broad range of applications. In order to account for both the actual solidification behavior of the material and to compare the results with published experimental microstructures from the system, two different parameter sets were used. For the first, the actual ternary eutectic composition was used, as indicated by E_{cal} in Fig. 1. To model the microstructure after solid state diffusion without the need to simulate the shift in solubility, a second parameter set was compiled. For this set, an off-eutectic composition is selected that produce the phase fractions comparable to those reported experimentally. This composition is depicted in Fig. 1 as E_{exp}.

This article is structured as follows: First we introduce the phase-field model and the evolution of the chemical potential. Then the applied framework and the implementation are introduced. The setup section describes the two parameter sets: the thermodynamic Calphad-database set and the experimental set. In the result section the patterns and their evolution during the simulations are presented. The simulations and an experimental micrograph are then compared, using nearest neighbor statistics. We conclude our paper by a short discussion of the results and the implications for further studies.

2. Model

To study the solidification process of a ternary eutectic system, we use a general thermodynamically consistent phase-field model, based on the work of [14–17].

2.1. Order parameter

For the model we consider the domain $\Omega \subset \mathbb{R}^d$ with $d \in \{1, 2, 3\}$, which is divided in $N \in \mathbb{N}$ subdomains $\Omega_\alpha \subseteq \Omega$, where each Ω_α is an order-parameter and represents a phase α, which is described by the positive part of the α phase-field, $\phi_\alpha : \Omega \times \mathbb{R} \to [0, 1]$, describing the local phase fractions at the position $\boldsymbol{x} \in \Omega$ and the time $t \in \mathbb{R}$. The function $\phi_\alpha(\boldsymbol{x}, t)$ is twice continuously differentiable. These phase-fields are defined as a vector on a simplex, $\boldsymbol{\phi} \in \Delta^N$, $\forall \boldsymbol{x}, t$ which is written as $\Delta^N = \{\boldsymbol{v} \in [0, 1]^N | \sum_{\alpha=1}^{N} v_\alpha = 1\}$, and represents the thermodynamic state of the phase.

2.2. The phase-field model

In order to model the total energy, we use the grand potential functional Ψ of the form

$$\Psi(\boldsymbol{\phi}, \boldsymbol{\mu}, T) = \int_\Omega \left(\varepsilon a(\boldsymbol{\phi}, \nabla \boldsymbol{\phi}) + \frac{1}{\varepsilon} \omega(\boldsymbol{\phi}) \right) + \psi(\boldsymbol{\phi}, \boldsymbol{\mu}, T) d\Omega \tag{1}$$

that consists of the gradient energy density $\varepsilon a(\boldsymbol{\phi}, \nabla \boldsymbol{\phi})$, the potential energy density $\omega(\boldsymbol{\phi})$ and the driving force $\psi(\boldsymbol{\phi}, \boldsymbol{\mu}, T)$. The functional depends on the order parameter $\boldsymbol{\phi}$, the chemical potential $\boldsymbol{\mu}(\boldsymbol{c}, T)$ and the temperature T.
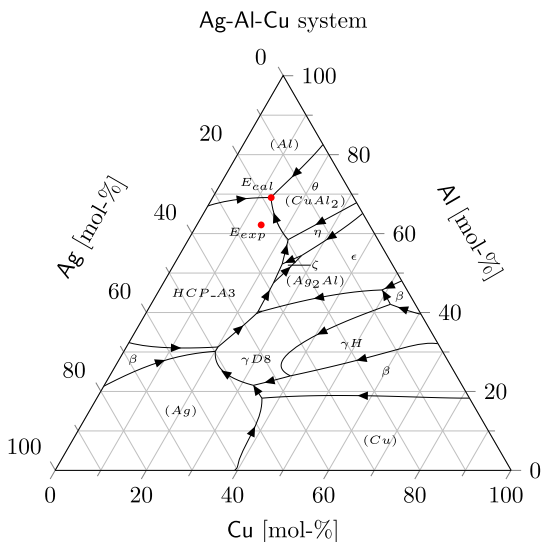

Fig. 1. Liquidus projection of the Ag–Al–Cu system based on the Calphad dataset. The thermodynamic ternary eutectic point is labeled as E_{cal}. The point E_{exp} was used to produce a simulation with phase fractions comparable to experiments [9–11].