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Spacing characterization in Al–Cu alloys directionally solidified under transient growth conditions

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Abstract

We study spacing selection in directional solidification of Al–Cu alloys under transient growth conditions. New experimental results are presented which reveal that the mean dendritic spacing vs. solidification front speed exhibits plateau-like regions separated by regions of rapid change, consistent with previous experiments of Losert and co-workers. Quantitative phase-field simulations of directional solidification with dynamical growth conditions approximating those in the experiments confirm this behavior. The mechanism of this type of change in mean dendrite arm spacing is consistent with the notion that a driven periodically modulated interface must overcome an energy barrier before becoming unstable, in accord with a previous theory of Langer and co-workers.

Keywords: Directional solidification; Dendrite; History dependent; Phase field simulation; Transient state

1. Introduction

Solidification microstructure is the starting point of any casting operation. Dendritic spacing and morphology established during casting often sets the scales of the downstream microstructure during manufacturing of alloys. This is particularly true in emerging technologies such as twin belt casting, where a reduced amount of thermomechanical downstream processing reduces the possibility of modifying microstructure length scales from that determined at the time of solidification.

Predicting columnar microstructure in cast alloys has been traditionally studied in the context of Bridgeman-type directional solidification conditions. Most studies have focused on the problem of primary and secondary arm spacing in dendrite arrays of thin liquid films of organic alloys, directionally solidified under steady-state cooling conditions, i.e. a sample is pulled through a constant thermal gradient at a constant pulling speed. Careful experiments on steady-state directional solidification reveal a reproducible correlation between spacing and pulling speed [1,2]. Studies of steady-state directional solidification have developed so-called geometric models to relate spacing to solidification processing parameters such as the pulling speed V, thermal gradient G and the alloy concentration c_o.

In geometric theories of spacing selection the structure and mathematical form of the dendrite arms is first assumed and a subsequent consistency relation is derived for the arm spacing (also referred to as "wavelength" in the literature). The construction and assumptions that go into setting up the geometry of the dendrite array lead to at least one phenomenological parameter that is fit to match the theory onto specific experimental spacing selection data [3–5]. While useful in helping to elucidate some aspects of spacing selection, such theories lack the fundamental element of microstructure predictability: the ability to self-consistently predict the morphology of the structure they are trying to say anything about. It is also not clear how such theories hold up to a change of conditions away from those of the experiments they were constructed to model.

While steady-state directional solidification is an important academic paradigm, it is not a realistic representation

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of the conditions prevalent during industrial casting, which normally occurs under rapidly changing growth conditions. Near a chill surface or, indeed, throughout the entire sample thickness in the case of very thin strips, the thermal gradient and solidification speed are neither constant nor independent of each other. Recognizing this, several research groups have attempted to extend geometrical models to include unsteady-state processing conditions. For example, Garcia and co-workers [6] and Kirkaldy and co-workers [4] have attempted to link the spacing of dendrite arms to the cooling rate $\dot{q}$. Once again, as with any geometric theory, phenomenological parameters are introduced to fit the model with experiments, although in this case the fits are not as good as in the steady-state case.

The importance of transient thermal processing conditions in establishing as-cast microstructures has been apparent since the theoretical work of Warren and Langer [7,8] and the experimental studies of Losert and Huang [9,10] on alloys of succinonitrile (SCN). Warren and Langer performed an analysis on the stability of dendritic arrays [7,8] and predicted that they remain stable to period doubling instability over a range of pulling speeds, contrary to the predictions of any geometric theory, whether steady-state or transient. Losert et al. [10] later observed that under a gradual change in pulling speed the spacing remained stable over a range of pulling speeds, consistent with the Warren and Lager predictions. They also noted the presence of an abrupt discontinuous jump in spacing at a particular pulling speed, which could be attributed to period doubling, also consistent with the predictions of Warren and Langer. In turn, Huang et al. [9] showed that by changing the rate of the pulling speed it is possible to obtain different spacings for a given set of final growth conditions. These works suggested a band of available spacings rather than a unique selection.

The results in the literature seem to point to two extremes. Under steady-state conditions, i.e. $dV/dt = 0$, the dendrite arm spacing appears to scale as a power-law of the pulling speed V, a result at least borne out qualitatively by geometrical models and some experiments. On the other hand, under transient solidification conditions, dendrite spacing and structure seem to depend strongly on transient history and initial conditions, at least in the idealized setting of a linear stability analysis or for well controlled experimental SCN dendrite arrays. The lack of unified theory to explain both these regimes likely points to an incomplete picture of the fundamental physics underlying microstructure selection in solidification. It also points to the need for a robust theory and modelling formalism that predicts the evolution of dendritic morphologies self-consistently, as function of only the input material parameters and cooling conditions, steady-state or transient.

Phase-field theory has emerged in recent years as promising candidate of a fundamental and self-consistent theory for modelling solidification microstructures. The first simulations to test spacing vs. pulling speed in alloys date back to the work of Warren and Boettinger [11], who found a monotonic band of spacings vs. pulling speed. The small system size used, however, precluded a quantitative comparison with experiments. Nowadays the phase-field methodology has become more quantitative by "marrying" simulations of phase-field models in the so-called thin interface limit [12,13] with novel simulation techniques like adaptive mesh refinement [14]. A first step using phase-field models to quantitatively model spacing in directional solidification was taken by Greenwood et al. [15] in two dimensions and Dantzig et al. in three dimensions [16]. These works modelled steady-state directional solidification in SCN alloys and found very good agreement (in the two-dimensional (2-D) limit) with the 2-D steady-state spacing experiments. These studies suggested that, at least under steady-state (i.e. Bridgeman growth) conditions and one type of initial condition (morphologically noisy initial interface), there could be a single crossover scaling function interpolating between the two power-law spacing regimes seen experimentally and modelled semi-empirically by geometrical models.

Despite the success of phase-field modelling in predicting steady-state spacing, as well as other steady-state properties such as cell tip structure [17], the methodology has not been used systematically to explore spacing under transient solidification conditions. Indeed, the ability to model cell, dendrite and seaweed structure, kinetic and surface tension anisotropy, different mobility, different thermal conditions and different initial condition makes phase-field modelling an ideal theoretical test ground to explore transient spacing development and how it may relate to the steady structures.

This paper reports new experiments and simulations that study primary spacing selection in directionally solidified Al–Cu alloys cooled under transient conditions closely related to those encountered in strip casting of Al alloys. The transient thermal gradient and interface speed are measured and correlated to measured dendrite spacing. Our results are shown to be inconsistent with steady-state and transient geometric theories. Instead, they suggest that there exists nearly stable ranges of spacings vs. front speed, connected by rapid changes in spacing at particular interface velocities. We also present new 2-D phase-field simulations that support this experimental picture. Our results are discussed in the context of Langer's theory as applied to a statistical distribution of spacings rather than a single unique spacing. In order to manage the length of this paper, a sequel to this paper [18] further explores some of the theoretical implications of the phase-field simulations presented here, focusing in particular on the connection between the transient behavior reported in this work and the steady-state behavior previously reported.

2. Experimental procedure

As-received Al–0.34 wt.% Cu samples were used to study solidification microstructure evolution under tran-