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Growth competition of columnar dendritic grains: A phase-field study

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Abstract—We report the results of an extensive phase-field study of the growth competition of columnar dendritic grains in two dimensions. We investigate the influence of the temperature gradient and grain bicrystallography on the selection of both grain and microstructure, focusing on a geometry with two grains with principal crystal axes oriented parallel and at a finite misorientation angle with respect to the axis of the temperature gradient. Our first main finding is that, for well-developed dendritic structures forming at a low-temperature gradient, the rate of elimination of the misoriented grain is a non-monotonic function of the difference in undercooling between the dendrite tips of the two grains. Hence this rate cannot be predicted even qualitatively by the common assumption that the elimination rate increases with this undercooling difference. The breakdown of this assumption is particularly striking for highly misoriented dendritic and degenerate structures that persist for very long times despite growing at a substantially larger undercooling than the well-oriented neighboring grains. Our second main finding is that microscopic thermal fluctuations at the origin of sidebranching can induce significant variations in the macroscopic trajectories of grain boundaries (GBs), thereby making grain selection a stochastic process, while yielding limited variations in the selected primary spacings. In contrast, in the absence of fluctuations, GB motion becomes essentially deterministic and grain elimination is suppressed. In addition, our simulations reproduce quantitatively scaling laws deduced from experiments for both the primary dendritic spacing and the dendrite growth direction of misoriented grains. They further reveal that the "intergrain" primary spacing selected by tertiary branching events at GBs is systematically larger than the "intragrain" primary spacing selected by the transient growth competition between primary branches within a single grain, while obeying the same scaling laws. Finally, the fact that the rate of grain elimination is slower in our 2-D simulations than in experiments suggests that the 3-D grain bicrystallography plays a key role in grain selection. This role is interpreted in the light of 2-D simulations that hinder sidebranching on the misoriented grain.

Keywords: Microstructure selection; Dendritic growth; Directional solidification; Phase-field method

1. Introduction

Most industrial solidification processes give rise to dendritic grain microstructures [1], which have a paramount influence on the mechanical behavior of materials [2–4]. Therefore, understanding the formation and evolution of grain boundaries (GBs) and the formation of new dendritic branches during dendritic growth is of major scientific and technological interest. Over several decades, transparent succinonitrile (SCN) alloys have been used as model systems to investigate solidification microstructure growth (e.g. [5–14]). Those experiments, combined with theoretical studies, have shed light on some fundamental aspects of dendritic growth, e.g. the sharp selection of dendrite tip radius [15–19], the wide range of stable primary dendritic spacing [20–24] and the selection of dendritic growth orientation [25–31] (for a comprehensive review, see Refs. [32,33]).

In comparison, the mechanisms of microstructure selection and GB evolution during polycrystalline dendritic growth is less clearly understood. Directional solidification experiments have provided insight into grain growth competition [10–12,14]. It was understood early that theories of polycrystalline dendritic microstructure selection need to account for the effect of sidebranching (e.g. [12]). At a GB, dendritic sidebranches may become new primary dendrites, making sidebranching the principal mechanism for the creation of new primary spacings, and hence for the selection of the inner grain structure. Furthermore, the outcome of the growth competition between sidebranches at a GB determines which of the two grains will occupy the liquid space in between the two crystals, and hence governs the shape of the resulting GB.

Dendritic sidebranching is a complex phenomenon, and systematic investigations of branching competition mechanisms in polycrystalline materials are still lacking. The origin of dendritic sidebranching is commonly accepted to be the selective amplification of thermal noise at the tip of the dendrite [34–40]. Once sidebranches reach a nonlinear regime, they grow competitively and this growth competition is largely deterministic [41,42]. However, the influence of these microscopic thermal fluctuations at the

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macroscopic scale, for instance in the selection of GB shapes, has to our knowledge never been discussed.

The classical model for competitive grain growth proposed by Walton and Chalmers [43] is mostly based on the fact that a misoriented dendrite grows at a higher undercooling than a dendrite oriented along the temperature gradient direction, and hence farther behind the solidification front. This minimum undercooling criterion has been commonly used to determine which microstructure is selected in the case of competitive columnar growth [44–46]. While early experiments seemed to agree well with this classical approach [10,11,45], later observations have shown that dendritic growth competition can be more complex [47–50]. For instance, misoriented grains can be the predominant origin of tertiary branches at a diverging GB, yielding a long-time coexistence of several different grain orientations [47,48]. Also, at a converging GB, misoriented primary dendrites have been observed to overgrow favorably oriented dendrites [50]. While Walton and Chalmers' theory only states that a dendrite misoriented with the temperature gradient cannot overgrow a more favorably oriented dendrite, the rate of elimination, i.e. the macroscopic orientation of the GBs, has remained relatively less explored. Experimental cross-sections have mostly exhibited GBs as straight lines [45,50] and the orientation of a diverging GB was suggested to follow a linear relation to the difference in misorientation of the two grains involved [50]. However, this latter observation was limited to low misorientation, i.e. 20° or less. Therefore, the common interpretation of the classical theory is that a higher crystal misorientation leads to a faster rate of elimination of the misoriented grain, even though some experiments show the long coexistence of grains of very different orientations [48].

In this study, we seek to discuss scaling laws for the selection of primary dendritic spacing and dendrite growth orientation, as well as the mechanisms of grain growth competition and selection at GBs, more specifically addressing the following questions:

- How similar is a primary dendritic spacing resulting from branching compared to one selected by coarsening/elimination?
- Does the primary spacing selected by dendritic branching in one grain depend on the orientation of its neighboring grain?
- What is the combined role of the temperature gradient and the grain orientations in the selection of GB orientation?
- Is a misoriented grain systematically eliminated faster if its misorientation is larger?
- Are GBs expected to develop as straight lines following a unique dynamic attractor, and is the GB orientation selection deterministic?
- What is the influence of the thermal fluctuations at the origin of dendritic sidebranching in the selection of macroscopic GB orientations and primary spacings?
- How does the transition from a dendritic to a degenerate structure affect the selection of GB orientations?
- How crucial is the role of the three-dimensional orientation of grains in dendritic grain selection?

In terms of computational methods, grain-scale simulations, e.g. using cellular automaton-based models [44,45], can predict some aspects of grain selection at the scale of macroscopic casting processes. They can, for instance, help design efficient geometries in order to produce single-crystal casting parts [51] or qualitatively reproduce the mechanisms of history-dependent selection of primary dendritic spacings [52]. However, these models are not detailed enough to investigate the specific mechanisms of dendritic branches competition at GBs, crucial in determining the macroscopic motion of GBs. Geometrically based branching models can also provide an estimate of the microstructural selection mechanisms (e.g. the orientation dependence of dendritic spacings [14]), but are not detailed enough to grasp the complex transient dynamics of branching competition.

At a smaller scale, phase-field (PF) modeling [53] has emerged as a powerful tool to study microstructure growth dynamics quantitatively. Its application to polycrystalline directional solidification has already highlighted complex selection mechanisms at converging GBs beyond the classical description by Walton and Chalmers [54], in agreement with experimental observations [50]. The long time scale survival of unfavorably oriented columnar grains has also recently been observed in a large-scale 3-D PF simulation [55].

In order to address the questions listed earlier in this introduction, we use quantitative PF calculations [56] to simulate the directional solidification of a bicrystalline SCN–acetone alloy. We perform numerous 2-D "numerical experiments", such as the one illustrated in Fig. 1, to build a systematic study of microstructure selection and growth competition mechanisms for two grains of different orientations upon directional solidification.

Throughout this article, the direction of the temperature gradient and the pulling velocity is chosen to be horizontal. The grain with a crystal orientation aligned with this direction is referred to as the favorably oriented (or well-oriented) grain and is represented in blue. The grain whose crystal orientation makes an angle α_0 with the temperature gradient direction is referred to as the unfavorably oriented (or misoriented) grain, and is represented in red. The GBs are referred to as converging or diverging if the primary dendritic tips on the two sides of the GB are respectively growing toward each other or away from each other.

Exploring a wide range of control parameters, namely the strength of the temperature gradient G and the crystal orientation α_0 (see labels in Fig. 1), we discuss the selection of (i) the primary dendritic spacings in the two grains, Λ_0 and Λ_α, (ii) the primary dendrite growth direction α, and (iii) the macroscopic (i.e. time-averaged) orientations of GBs θ_C and θ_D, as well as the possible fluctuations in these selection mechanisms.

In Section 2, we review existing theories for microstructure selection mechanisms, which we discuss later in Section 4 in light of results from the PF simulations presented in Section 3.

2. Microstructure selection theory

2.1. Primary dendritic spacing

Several experiments (e.g. [21,22,24]) and numerical studies (e.g. [52,57,58]) have shown that a wide range of stable primary dendritic spacing Λ may be selected from similar