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In situ observation of solidification patterns in diffusive conditions

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ABSTRACT

We present a review of recent in situ experimentation studies of solidification front patterns and microstructures in alloys. Front-tracking diagnostics and real-time observation methods using high-resolution optical or X-ray imaging devices currently apply to model transparent systems as well as metallic alloys in thin and bulk samples. On a theoretical basis that spans the physics of nonequilibrium pattern formation and materials science, in combination with time-resolved numerical simulations, conclusive results of both fundamental- and applied-science interest have been obtained on major problems relative to multiscale microstructure selection, morphological transitions, and crystallographic effects during single- and multi-phase solidification. We will mainly focus on the dynamics of cellular, dendritic, and eutectic growth patterns in diffusive-growth conditions, that is, in the absence of convection in the liquid. This can be achieved in (semi-)thin samples, or, for bulk solidification, in the reduced-gravity environment of orbiting facilities. A selection of emerging work on, e.g., faceted growth and adaptive control of solidification patterns will furthermore be reported. We conclude by pointing out open questions and new perspectives for future research.

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1. Introduction

The prediction and control of solidification microstructures in alloys actively stimulates abundant laboratory research linking fundamental and applied sciences. The advancement of in situ experimental methods for real-time observation of the propagating solid–liquid interface during solidification is key to this vast scientific enterprise. Solidification microstructures –modulations of chemical composition, regular arrangements of finely dispersed crystal phases, and lattice defect networks in polycrystals– are a trace, left frozen in the bulk solid, imprinted by growth front patterns. In metals, and nonfaceted materials in general, these self-organizing phenomena are essentially determined by the redistribution of heat and chemical species by diffusion –often “assisted” by convection motions in the melt– and by local equilibrium or fast attachment kinetics at the interfaces. The dynamical origin of solidification microstructures has long been attested [1–3], and their complexity more and more clearly revealed over time by metallurgical studies, but a consistent nonlinear-physics approach of nonequilibrium pattern formation phenomena has been fostered only in the 1980's [4–7]. During the last two decades, systematic experimental studies have been undertaken in a coherent theoretical context, implementing high-resolution front-tracking diagnostics and real-time observation methods in polycrystals and multiphase composites on the required time and space scales. In parallel, time-resolved numerical simulations quickly developed [8–10]. By these combined means, conclusive results have been obtained with unparalleled accuracy on major questions concerning morphology selection and transition processes, microstructural defects, and crystallographic effects during one- and multi-phase solidification, thus opening up to further prospects on both theoretical and technological levels.

We present a review of recent experimental results obtained by real-time observation methods during solidification of dilute and eutectic alloys. Major breakthroughs from *in situ solidification* (shortly speaking) research can be traced back to the implementation, in the 1960s, of thin-sample directional solidification of transparent alloys “that freeze like metals” by Jackson and Hunt [11,12], and further developments during the 1970–1980 decades [13] (also see Ref. [14]). Since the middle of the 1990's, discoveries have followed an accelerated pace. Real-time observation has been extended to bulk transparent alloys during directional solidification, and, by taking advantage of high-brilliance X-ray facilities, to metallic alloys. For clarification, we will consider situations where perturbing factors coming from gravity- (or capillarity-) driven convection in the liquid were minimised, thus closely approaching the ideal limit of diffusion controlled growth. We also deliberately place ourselves in conditions close to conventional casting, therefore excluding fast solidification from our report. Focus will be put on experimental studies based on directional-solidification methods performed with thin samples, and, for bulk solidification, in reduced-gravity conditions in orbiting facilities. We will highlight the success of a synergic research combining in situ solidification and numerical modelling, which is highly exemplary, and particularly fertile. In these conditions, the morphological stability of dendritic, cellular, and lamellar/rod-eutectic solidification patterns have been carefully tested. Moreover, remarkable growth shapes (doublon, spiral two-phase dendrite) and complex space-time phenomena in drifting and oscillatory patterns have been discovered. The strong history and boundary-condition dependency of extended solidification patterns has been clearly demonstrated. New insights into initial and transient stages, as well as mild instrumental forcing (thermal bias, isotherm curvature) have been gained. Our understanding of the interplay of the solidification dynamics with crystal-lattice defects, and with the crystallographic anisotropy of the solid–liquid and solid–solid interfaces involved in the solidification process is also deeply improving. Most of the overviewed studies share in common a general-physics approach of solidification front pattern formation and stability, and are directed towards the unraveling of complex aspects of the nonlinear dynamics involved in the formation of solidification microstructures. They also openly keep a tight link with pending questions of great importance in materials science (e.g. columnar *vs* equiaxed growth, polycrystal and eutectic-grain textures), from which they are obviously inspired, and on which they cast new light. This corpus of experimental findings is of essential help for validating fundamental models and existing predictive codes. It should moreover provide a new basis for optimising the modelization of natural and industrial solidification processes, during which fluid flow in the liquid and plastic deformation in the solid are involved.

This review is structured as follows. We briefly present the general context (diffusion controlled growth experiments in thin samples and in microgravity) in Section 2, and novel observation methods in Section 3. The main results are presented in the following order. Section 4 is dedicated to dendritic and cellular patterns in dilute alloys. It is divided into three parts. In Section 4.1 (thin-sample solidification), particular emphasis will be put on the role of interfacial anisotropy in the stability of dendritic (Section 4.1.1) and cellular (Section 4.1.2) patterns. The dynamics of grain-boundary grooves (Section 4.1.3), and a novel method developed for a local adaptive control of solidification patterns (Section 4.1.4) will be briefly presented. In Section 4.2, we will focus on bulk solidification in nearly diffusive conditions, that is, in practice, in microgravity (μg) facilities. After a brief report on pioneering work (Section 4.2.1), optical observations of cellular and dendritic arrays in transparent alloys (Section 4.2.2) will be described. The presentation of X-ray radiography studies of columnar and equiaxed growth regimes in metallic samples (Section 4.2.3) opens up to a brief discussion on possible mechanisms (including convection or advection motions in the interdendritic liquid) at play in dendrite-arm fragmentation processes. In Section 4.3, some prospective studies on faceted growth –in particular, polycrystalline silicon– will be mentioned. Eutectic growth is presented in Section 5. It will

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