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Eutectic solidification patterns: Interest of microgravity environment

Structures de solidification eutectique : de l'intérêt d'un environnement de micropesanteur

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The solidification of binary eutectic alloys produces two-phase composite materials in which the microstructure, that is, the geometrical distribution of the two solid phases, results from complex pattern-formation processes at the moving solid–liquid interface. Since the volume fraction of the two solids depends on the local composition, solidification dynamics can be strongly influenced by thermosolutal convection in the liquid. In this contribution, we review our experimental and numerical work devoted to the understanding of eutectic solidification under purely diffusive conditions, which will soon be tested and extended during the microgravity experiment TRANSPARENT ALLOYS planned by the European Space Agency (ESA).

R É S U M É

La solidification des alliages eutectiques binaires produit des matériaux composites biphasés, dont la microstructure, c'est-à-dire l'arrangement géométrique des deux phases cristallines dans le solide, résulte d'un processus complexe d'auto-organisation à l'interface solide–liquide en cours de croissance. Puisque la fraction volumique des phases solides est une fonction de la composition locale, la dynamique de solidification peut être fortement influencée par des mouvements de convection thermo-solutale dans le liquide. Dans cet article, nous faisons le point sur nos travaux expérimentaux et numériques dédiés à la compréhension de la croissance eutectique en conditions de transport purement diffusif. Ces résultats seront bientôt testés et étendus dans une expérience en micropesanteur TRANSPARENT ALLOYS prévue par l'Agence spatiale européenne (ESA).

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1. Introduction

Solidification phenomena have played an important role in the history of human civilization, to the point that various epochs are named after solidified materials. Our remote ancestors fought against harsh climatic conditions during the ice age. During the bronze age and the iron age, the mastery of increasingly complex metallurgical techniques was a key ingredient for progress. Later on, the industrial revolution was largely based on the capacity to produce steel in large quantities. Finally, the development of microelectronics was only possible thanks to the progress of solidification-based purification techniques (e.g., zone-refinement) and single-crystal growth for silicon. Thus, solidification has accompanied the progress of human civilization since the dawn of times.

There have also been various links between solidification and geographic exploration. One of the starting points of modern solidification science is the work of Stefan, who formulated in 1889 the mathematical model to describe the growth of an ice layer on water in contact with cold air [1,2]. The field data against which Stefan tested his theory came from several polar expeditions [3]. With various extensions, this type of mathematical formulation has been extensively used later on to describe growth processes, and mathematicians often refer to such free-boundary problems as “Stefan problems” [4].

While the race for the poles, and the struggle to reach the most remote points on the planet, were at the forefront of discovery at the end of the 19th century, today, so to say, space is the final frontier. Quite curiously, there is again a link between exploration and solidification. One of the major assumptions in Stefan’s theory was that heat transport takes place by conduction only. This is true in the setting considered in Stefan’s original work, where heat conduction takes place in the solid ice. However, as soon as heat transport is driven by temperature gradients in the liquid, the resulting density variations trigger convective fluid motion. Density variations are even stronger in crystal growth from a multicomponent liquid mixture, where both heat and chemical components need to be transported. In fact, diffusive conditions during solidification can be realized on earth only in samples in which at least one dimension is in the 10–100 μm range, such that viscous friction at the walls prevents or reduces the fluid motion.

The access to space, and thus to an environment with reduced gravity levels, has offered the possibility to study solidification in macroscopic samples without, or at least with a low level of convection. Solidification experiments in sounding rockets, space vehicles or space stations have a long tradition [5–15]. The main goal of such experiments is to provide reference data and observations against which modern theories and models of solidification can be tested and validated for further refinement and use under terrestrial gravity conditions. As a consequence, contrary to the historic example of the Stefan problem, in which a theory was developed only long after the field data had been retrieved, today modeling accompanies solidification science both in the prematuration of new space experiments and in the interpretation of data obtained in microgravity.

Solidification is a classical example of pattern formation outside of equilibrium, in which a structured final state forms by self-organization processes from a structureless initial state [16,17]. The solidification of metallic alloys results in the spontaneous development of a large variety of *microstructures* [18]: dendrites, cells, multi-phase eutectic or peritectic composites, and multi-scale architectures that are made of combinations of several subunits. These structures are a frozen trace of interfacial patterns that exhibit a complex spatio-temporal dynamics. Although solidification is only the first stage of materials fabrication, and is often followed by heat treatments, rolling, or other processing steps, the material usually keeps a memory of the initial structuration, and therefore the solidification microstructures largely influence materials properties. For a physicist, it is a challenge to understand by which mechanisms these structures form, and how their genesis can be controlled and guided towards desirable target structures.

The study of solidification science from the viewpoint of the modern theory of pattern formation started in the 1980s [19] and has been ongoing ever since. The fundamental processes that shape the microstructures are well known: structures result from a subtle interplay between the transport of heat and chemical components, and the properties of the solid–liquid interfaces. For nonfaceted crystal growth, the most important interfacial effect is the Gibbs–Thomson effect, which originates from capillarity. Since these processes occur at the moving interface of a growing structure, both in experiments and numerical simulations it is necessary to follow the *dynamics* of the solidification front, and access the time evolution of the entire structure. In experiments, this requires an observation *in situ* during growth; in numerical simulations, it is necessary to solve the complete free-boundary, or Stefan problem. A close comparison between experiments and simulations is crucial, both to validate the mathematical and numerical models and to understand the fine details of the pattern formation process.

The French space agency CNES has set up a long-term fundamental research program on solidification in microgravity environments, which supports ground experiments as well as numerical simulations to prepare experiments in space. Here, we will review some aspects of our joint work on the solidification of eutectic alloys. In order to boldly go where no one has gone before we have combined innovative in situ experimental techniques with numerical simulations that use state of the art phase-field methods.

The remainder of this paper is structured as follows. We summarize the general background of our research in Section 2, and briefly introduce our experimental and numerical methods in Sec. 3. In Sec. 4, we review our explorations of patterns beyond simple lamellae and rods, and in Sec. 5 we examine the effects of “deformations” of the thermal field on eutectic solidification patterns. A further discussion of several aspects (Sec. 6) is followed by conclusions and perspectives in Sec. 7.