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Wave dynamics on directional solidification interfaces swept by a flow in a thin sample

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ABSTRACT

The effects of a transverse flow on the dynamics of a directional solidification interface are studied experimentally in a thin sample. The set-up enables a non-intrusive visualization of the interface and an independent control of both the flow and the solidification conditions. The flow is forced in the sample from an external thermosiphon which provides an accurate steady velocity up to 1.2 mm/s. A transparent melt of succinonitrile is used with a sample depth allowing the solidification of a single layer of microstructures. Downstream inclinations of microstructures and downstream promotion of dendritic sidebranching are observed. Surprisingly, large scale traveling waves involving a wavelength of several cells or dendrites are evidenced on the interface in a large range of conditions. Two kinds of waves are evidenced, one involving a slow velocity, a weak amplitude and a sinusoidal profile, the other a large velocity, a large amplitude and a non-linear profile. Both result from the coupling between solidification and flow and induce striations in the solid phase.

1. Introduction

Solidification of a melt in a thermal gradient usually goes together with the generation of flows by thermal or thermosolutal convection. In addition, some flows may be forced by external means, for instance by seed rotation in Czochralski process [1] or by magnetic stirring [2]. These flows then mix the liquid medium and may even yield dendrite fragmentation and transport of seeds away from the interface. Altogether they thus largely participate to the history of the structure and the composition of the resulting solid and have accordingly received considerable attention. However, another possible effect of flows on solidification is the generation of new instabilities by a coupling between a flow and a solidifying interface. In particular, as the solid phase imposes a boundary condition to the flow, the deformation of the solidifying interface may modify the flow which in turn may interfere with the interface dynamics by solute advection. On this ground, a possible stabilization of planar interfaces by flows has long been predicted for some directions of perturbations and flow geometries [3] and has received some support from experiments [4–6]. Farther from the onset of planar destabilization, a direct effect of flows on microstructure has been investigated on microstructure inclination and on the asymmetry of dendritic sidebranching. However, no coupled instability for a set of growing microstructures and a flow seemed to have been predicted or evidenced so far.

Here, our objective has been to carefully study the dynamic coupling between a directional solidification interface and a flow in well controlled conditions. For this, we used a thin sample in which natural convection is inhibited by dissipation due to the large viscous stresses displayed at the plates that sandwich the melt. This way, only forced flows could be induced with no opportunity of natural perturbation. In addition, thin samples allow a direct, non-perturbative, visualization of the solidification interface of transparent melts and thus an easy detection of dynamical events. Here, we used a transparent succinonitrile melt and flows externally generated by a thermosiphon. Altogether, this enabled us to evidence the generation of interfacial waves by coupling between flow and solidification interface [7]. Two kinds of waves have been distinguished and their main features have been documented. Both yield the generation of periodic modulations in the solid phase that resemble the striations sometimes observed in cast materials.

In the following, we first detail in Section 1 the experimental set-up used in this study and in Section 2 its specificity regarding the onset of planar instability. We then report in Section 3 on the effects of flows on microstructure inclinations and on sidebranching asymmetry. In Section 4, we address the main result of this

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study, the evidence of flow-induced interfacial waves, and we document their main features. We finally discuss the relevance of these results and conclude about this study.

The experimental set-up is designed to achieve directional solidification in homogeneous and controlled conditions [8–10] together with a permanent visualization of the interface dynamics. To this aim, solidification proceeds in thin samples that are filled with a transparent material and whose depth corresponds to the typical thickness of a layer of microstructures. Samples are then pushed at a controlled speed in between heaters and coolers that set a uniform thermal gradient (Fig. 1a). Heaters and coolers are made of top and bottom metallic blocks that are electronically regulated at 100 °C and 10 °C respectively to an accuracy better than 0.1 °C . They yield a thermal gradient G of either 70 K cm^{-1} or 140 K cm^{-1} on the solidification interface, depending on the gap imposed between the blocks by suitable spacers. The pushing stage is provided by a micro-stepper motor which drives a linear ball-screw attached to a translating stage. Pushing velocities up to 50 μm s^{-1} may be achieved with a relative accuracy better than $\pm 3\%$ on long duration.

Samples consist of two glass plates separated by spacers. They are filled with the mixture to solidify, here a dilute alloy of succinonitrile with acrylonitrile as solute. As the melting temperature of succinonitrile is 58 °C when pure, the solidification interface sets around the middle of the gap between heaters and coolers. This ensures that the sample translation negligibly modifies the thermal gradient on the interface [11]. Samples are wide (4.5 cm) and long (15 cm) enough to provide solidification domains far from boundary disturbances. Their thickness was chosen small enough to avoid the emergence of a second layer of microstructures and large enough to provide a 3D behavior of microstructures. This ensured that microstructures behave similar to those of a multi-layer solidification, in particular regarding forms [12,13], undercooling [12,14], sidebranching [8] or instabilities [15]. Thanks to the transparency of the filled sample, visualization of the solidification interface is achieved from the slight optical aberrations undergone by a parallel light beam crossing the sample, which reveal the interface as a sharp contrast line. The corresponding images are recorded by a camera involving 1024 × 768 pixels. In the grooves, images sometimes show dark channels which correspond to a gaseous phase that does not couple with the interface dynamics.

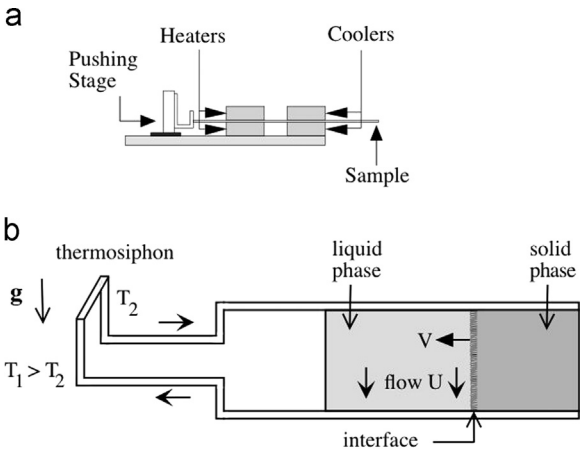

Fig. 1. (a) Sketch of the usual solidification set-up. A motor pushes a sample within a thermal gradient induced by heaters and coolers. Observation is achieved by ombroscopy on a parallel light beam crossing the sample at the interface location. (b) Thermosiphon added to force a controlled flow U in the sample. This flow sweeps the solidification interface and induces, above a growth velocity V, a coupled solidification-hydrodynamic instability that yields traveling waves and striations (Fig. 3).

To improve the microstructure homogeneity, care has been taken to prepare the sample in a single crystal state. This has been achieved by making a selected grain invade the whole sample by a spatial control of fusion/solidification prior to the sample insertion in the experimental set-up. The chosen orientation was such that the principal axes of the cubic crystal of succinonitrile were aligned on the thermal gradient direction, the mean interface direction and the cell depth. Control of the orientation was achieved from the symmetry displayed by the sidebranches of freely growing germs and of dendrites rapidly growing in a thermal gradient.

An interesting feature of thin samples is to inhibit natural convection owing to the large viscous dissipation imposed by their small depth. This situation, which forbids the occurrence of spurious uncontrolled flows, is thus especially suitable for achieving a control of flows. It however requires forcing flows either mechanically by moving solid parts or hydrodynamically by injecting flows. Here, we preferred using the latter mean owing to the difficulty in controlling mechanical parts in a thin sample. In addition, we looked for a permanent continuous injection in contrast with the discrete injections used in [6]. This was provided by connecting the sample to an external thermosiphon that is known to induce low amplitude controlled flows in similar configurations [16]. It consists in two vertical branches that are differentially heated so that the fluid is lighter in one vertical branch than in the other. These branches are then connected to each side of the sample by flexible tubes to yield a closed circuit. They then provide a hydrostatic pressure difference between the two sides of the sample that sets the fluid into motion in between. Within the sample, the flow is then parallel to the solidification interface (Fig. 1b) and, following the small sample depth d, it stands in a Stokes regime with a Reynolds number $Re = dU/\nu$ of order 10^{-3} in our range, ν denoting the fluid kinematic viscosity and U the flow amplitude. It then displays a Poiseuille profile.

A simple modeling of the thermosiphon can be obtained from the Navier–Stokes equation in the Stokes regime by considering its circulation along a streamline.

$$-\oint \nabla p \, d\mathbf{l} + \oint \rho \mathbf{g} \, d\mathbf{l} + \oint \mu \Delta \mathbf{v} \, d\mathbf{l} = 0 \tag{1}$$

where p denotes the fluid pressure, ρ the fluid volumic mass, μ the fluid dynamic viscosity, $\mathbf{g}$ the acceleration of gravity and $\mathbf{v}$ the fluid velocity. The circuit being horizontal, except in the thermosiphon, the contribution of gravity reduces to the differential effects of thermal dilation in the thermosiphon branches. Following the Poiseuille flow profile, the contribution of dissipation is proportional to the constant net mass flow D. The proportionality factor, called the hydrodynamic resistance R_h, depends on the geometry of the cross section. As the circulation of the pressure gradient vanishes, one finally obtains :

$$\rho_0 \alpha \Delta T g H = D \sum_i R_{h,i} \tag{2}$$

where ρ_0, α, H and ΔT are the fluid volumic mass at the freezing temperature, its thermal dilation factor, the length of the thermosiphon branches and their temperature difference. The index "i" labels the different parts of the closed loop and $R_{h,i}$ stands for their hydrodynamical resistance. In particular, for a cylindrical section of length l_c and diameter D_c, one gets $R_h = 128/\pi \mu l_c D_c^{-4}$ and for the rectangular liquid domain ahead of the interface of length L, width l and depth d, $R_h = 12\mu d^{-3} L^{-1} l$. The net mass flow D and thus the flow amplitude U are then directly proportional to the temperature difference between the thermosiphon branches. This ensures their accurate tune, independent of the growth conditions. With a maximum temperature difference of $\Delta T = 32$ °C, one obtains this way a maximal flow amplitude ranging experimentally from 300 μm s^{-1} at $d = 150$ μm to 1200 μm s^{-1} at $d = 400$ μm. We

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