INFLUENCE OF AN EXTERNAL PERIODIC FLOW
ON DENDRITIC CRYSTAL GROWTH

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1 - INTRODUCTION

The problem of dendritic shape selection in crystal growth processes has been the subject of many theoretical and experimental studies these last years(1-8). Experimentally, dendritic side-branching has received less attention despite the significant number of theoretical and numerical studies devoted to this problem(9-12). A recent experimental study(13) shows results consistent with the assumption that side-branching originates into the selective amplification of microscopic noise at the crystal tip. Indeed, for testing such an assumption, it would be necessary to control the noise in the experimental system. Such a control has been achieved in the case of anomalous Saffman-Taylor fingers(14). In the present crystal growth experiment, we achieve a control of the side-branching by imposing a time dependent flow during the growth process. We report herein a first quantitative study of the effect of a periodic external flow on dendritic growth.

2 - EXPERIMENTAL SETUP

The system from which crystals grow is a mixture of pivalic acid and ethanol alcohol (PVA/AL). The crystallographic structure of the solid phase (which is essentially pivalic acid) is fcc(4). In all the experiments, the alcohol concentration is 0.7 \% in volume. This corresponds to a melting temperature for the mixture of 25 °C. Because of the presence of alcohol which plays the role of impurity, the growth is controlled by chemical diffusion.

The quartz cell is a parallelepiped of dimensions 45 x 5 x 1 mm. In order to obtain a longitudinal flow of the PVA/AL system, an open hydraulic circuit is formed, including the cell, small diameter tubes and a reservoir. The time dependent flow is produced by varying periodically the position of the reservoir, at a frequency $v_{\text{exc}}$ ranging between 5 and 500 mHz. The maximum velocity amplitude of the resulting flow is fixed at the value 19.6 μm/s for all the experiments ; there is no mean flow.

In a similar way as in a previous work(15), the cell is entirely immersed in a circulation of thermally regulated water maintained at temperature $T_1$ ; by this way, a regulation better than 20 mK for the operating temperature is achieved. On the other hand, the tubes forming the hydraulic circuit are in thermal contact with another circulation of thermally regulated water ; such tubes can therefore be held at a temperature $T_2$ slightly above $T_1$. Most of the experiments are done with an overheating $\Delta T = T_2 - T_1 \approx 0.2°C$, which is sufficient to avoid cristallization in the tubes and small enough to leave the growth process unperturbed by any temperature gradient effect.

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The dendrites are observed with a microscope which allows magnification up to 750. The images are sent to a video camera and digitized into 512 x 512 pixels. The resulting overall image separation ranges from 0.6 to 0.9 µm/pixel.

3 - EXPERIMENTAL PROCEDURE

The experimental procedure is as follows: starting from a crystallized state, the cell is heated up at temperature $T_1$ slightly below equilibrium temperature $T_e$ so that only a single crystal remains in the cell. Then the periodic flow is turned on and after a few minutes the cell is cooled down at a temperature $T_o$ smaller than $T_1$ by an amount ranging from 0.3 °C to 1 °C. After a short transient of a few minutes (which corresponds to the thermal time constant of the system), dendrites grow from the germ in the <100> directions. We choose one isolated dendrite and follow its temporal evolution. In all the experiments, the flow is directed along the growth direction of the dendrite.

4 - RESULTS

A typical dendrite obtained in the presence of an external periodic flow is shown in Pic. 1. In this case, the amplitude of the flow is 19.6 µm/s, the imposed frequency 15 mHz, the growth velocity 0.9 µm/s and the tip radius 8.0 µm. Also shown, on the lower right of the picture, a "natural" dendrite of pivalic acid, i.e. a dendrite growing out of a solution at rest. One obtains, when the periodic external flow is applied, a spectacular regularity of the spatial structure of the side-branching. The wavelength of the side-branching is well defined in this case, and the branches appear to be correlated over large distances – typically one hundred times the radius of curvature at the tip –; this contrasts with natural dendrites where coherence rapidly decays as the distance from the tip increases(13). The apparent wavelength of the side-branching can be estimated to 65 µm in the case corresponding to Pic. 1. This is close to the imposed wavelength $\lambda_{exc.} = \frac{V}{v}$, which is about 60 µm in this case, so that one can consider that the actual wavelength of the side-branches is imposed by the forcing.