Section 1: Single crystal materials

# NUMERICAL SIMULATION OF THE FLOW FIELD IN SHAPED CRYSTAL GROWTH PROCESS

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The purpose of this study is to clarify the effect of hydrodynamics on the bubble generation and repartition in shaped sapphire. The influence of the Reynolds numbers (in the meniscus and in the capillary channel) and of the Marangoni number on the flow field was studied with the help of numerical simulation. The governing equations (of mass and momentum conservation) and also the geometrical and physical parameters have been nondimensionalised. A strong influence of the Reynolds numbers in the meniscus (Re<sup>men</sup>) and in the capillary channel (Re<sup>cap</sup>) have been established. The velocities in the liquid decrease when Re<sup>men</sup> increases and the maximum velocity position moves from the left of the exit of the capillary channel to the axis of symmetry. This behaviour is independent of the  $Re^{cap}$ . A critical value of Re<sup>men</sup> was found for which the maximum velocity remains constant. The surface tension variation, leading to Marangoni effect, strongly influences the flow field. For a meniscus height of 100µm the apparition of a loop in the flow field due to the surface tension variation was observed for Marangoni numbers (Ma) greater than a critical value (~1.5). Till this value the velocities are comparable to the pulling rate except a small region close to the meniscus surface where velocities are vanishing. The dimensions of the region above mentioned is comparable to the meniscus height. In this region it is possible that the gas contained in the liquid or the micro-bubbles accumulate till they are incorporated into the crystal. The maximum velocity of the meniscus surface shows a linear variation with the Marangoni number. For a constant Marangoni number and for different Reynolds numbers in the meniscus it was observed that the velocities composing the loop increase with the increase of the meniscus height.

Keywords: Sapphire, Micro-bubbles, Hydrodynamics, Shaped crystals

## 1. Introduction

A lot of variants of the Stepanov techniques have been developed in order to obtain shaped sapphire. However, these techniques generally lead to the presence of micro-bubbles in the grown crystals [1-4].

Among the various phenomena involved in the apparition and incorporation of bubbles in the growing crystal, the flow of the liquid, from the crucible to the solid-liquid interface, is of major importance because it is supposed to carry the dissolved gases and the bubbles. The aim of the present work is to study, with the help of numerical simulation, the flow field in the capillary die and the effect of the modification of the leading parameters: geometry and growth rate. The influence of the surface tension variation, leading to Marangoni convection on the liquid meniscus [5], has also been analysed.

## 2. Numerical simulation

We consider the growth of axi-symmetric sapphire rods [6]. The numerical simulation of the physical phenomena involved in the liquid, including the equations of momentum and mass conservation (2.1 and 2.2), has been perform with the help of the FIDAP finite elements software.

$$\rho \cdot \mathbf{u}_{j} \mathbf{u}_{i,j} = -\mathbf{p}_{,i} + \eta \cdot \left[\mathbf{u}_{i,j}\right]_{,j}$$
(2.1)

$$u_{i,i} = 0$$
 (2.2)

The fluid flow is considered in a steady state because the pulling rate and the fluid velocities are in the order of mm/min and the length of the crystal is decimetric. The process is laminar because the low fluid velocities and pulling rate determine small values for the Reynolds numbers. The fluid is newtonian and incompressible.

The configuration is axy-symmetric and limited to the liquid in the capillary die and in the meniscus. The geometry and the boundary conditions for velocities are given in Fig. 1. The geometry of the meniscus and of the solid-liquid interface are taken from experimental parameters [7].

The geometrical and physical variables are in a number of six: the pulling rate  $(v_T)$ , the meniscus height (h), the crystal radius ( $R_{cryst}$ ), the capillary die radius ( $R_{cap}$ ), the density of the liquid material ( $\rho$ ) and the viscosity of the melt ( $\mu$ ).

In order to adimensionalise [8] the equations of momentum and mass conservation (2.4 and 2.5) the radius of the capillary die and a velocity which is proportional to the pulling rate are introduced as characteristic values:

$$U = \frac{S_{cryst}}{S_{cap}} \cdot v_{T}$$
(2.3)

$$u_{j}^{*}u_{i,j}^{*} = -p_{,i}^{*} + \frac{1}{Re}\tau_{ij,j}^{*}$$
(2.4)

$$\mathbf{\mu}^*_{j,j} = 0$$
(2.5)

where  $S_{cryst}$  is the section of the crystal,  $S_{cap}$  is the section of the capillary and  $v_T$  is the pulling rate.



Fig. 1. Geometry and boundary conditions.

In order to reduce the number of variables, the nondimensionalisation of the problem led to the four main variables:

- A: the ratio of the radius of the crystal to the radius of the capillary die,  $A = \frac{R_{cryst}}{R_{cap}}$ .

-  $R_e^{cap}$ : the Reynolds number in the capillary channel,  $Re^{cap} = \frac{\rho}{\mu} R_{cap} \cdot U$ .

-  $R_e^{\text{men}}$ : the Reynolds number in the meniscus,  $Re^{\text{men}} = \frac{\rho}{\mu} \mathbf{h} \cdot \mathbf{v}_T$ .

- Ma: the Marangoni number,  $Ma = \frac{|\dot{\gamma}_T \cdot \Delta T|}{\mu \cdot U}$  where  $\dot{\gamma}_T$  is the variation of the surface tension with

temperature and  $\Delta T$  is the temperature difference between the top and the bottom of the meniscus.

In the Table 1 the physical and geometrical variables, the nondimensionalised variables and the values of these variables as a function of the four parameters above mentioned are given.

Table 1. Geometrical and physical parameters.

Geometrical & physical	Physical symbol	Nondimensional symbol	Value
Pulling rate	Væ	v_*	$1/\Delta^2$
Meniscus height	h	h*	$(\text{Re}^{\text{men}}/\text{Re}^{\text{cap}}) \text{A}^2$
Crystal radius	R <sub>crvst</sub>	R* <sub>crvst</sub>	A
Capillary die radius	R <sub>cap</sub>	R* <sub>cap</sub>	1
Density	ρ	1	1
Viscosity	μ	1/Re <sup>cap</sup>	1/Re <sup>cap</sup>

The iterative method used to obtain the nonlinear steady solution was the successive substitution (S. S.). Function of the meniscus height the numbers of the mesh nodes is between 9849 and 12425.

## 3. Results and discussion

The influence of the Reynolds numbers on the flow field in the meniscus and in the capillary channel has been studied. It was also observed that the Marangoni number influences the distribution of the velocities at the boundary of the meniscus.

#### 3.1. Influence of Reynolds numbers on the flow field

In order to show the influence of Reynolds numbers in the meniscus ( $Re^{men}$ ) and in the capillary channel ( $Re^{cap}$ ) on the flow field, realistic experimental situations were considered. The minimal and maximal limits of the two Reynolds numbers were chosen in order to have the height of the meniscus between 60 and 350µm. Such meniscus heights have been measured by Theodore [7] for shaped sapphire growth. Fig. 2 shows the flow field as a function of  $Re^{men}$  for a constant value of  $Re^{cap}$ . The meniscus height increases when  $Re^{men}$  increases. The velocities in the liquid decreases when  $Re^{men}$  increases. For low values of  $Re^{men}$  the maximal velocity in the liquid is situated on the left of the exit of the capillary channel, in the meniscus. This case corresponds to a minimum value of the meniscus height (60µm). When  $Re^{men}$  increases the maximum velocity of the liquid moves to the centre of the geometry, but still in the meniscus (Fig. 2b and c) or to the center of the geometry on the axis of symmetry and in the capillary channel (Fig. 2d and e).



Fig. 2. Flow field for Re<sup>cap</sup>=0.02.

Fig. 3 shows the variation of the maximal velocity as a function of  $Re^{cap}$  and  $Re^{men}$ . The maximum liquid velocity decreases when the Reynolds number in the meniscus increases. At the beginning, the decrease is almost linear till a critical value of  $Re^{men}$  is reached. Then the maximum velocity is constant. However, this critical value corresponding to a meniscus height of 400µm for sapphire, is not reached experimentally (h<350µm).

The maximum velocity of the liquid increases a lot with the increase of the Reynolds number in the capillary channel ( $Re^{cap}$ ) but the variation of the maximum velocity with  $Re^{men}$  is the same for all values of  $Re^{cap}$ .



Fig. 3. Maximum velocity in the liquid as a function of Re<sup>cap</sup> and Re<sup>men</sup>.

### 3.2. Influence of the Marangoni effect on the flow field

On the meniscus external surface (Fig. 1) the temperature is not constant. This fact determines a variation of the surface tension. The nondimensionalisation gives a value of the surface tension:

$$\sigma_{ij}^* = \frac{\sigma_{ij}}{\rho \cdot U^2} \tag{3.2.1}$$

In order to calculate the surface tension effect the thermal gradient was chosen in the following experimental interval: (5K/mm-100K/mm). The nondimensional parameters were chosen in order to get constant meniscus heights. The following results are given for a meniscus height of 100 $\mu$ m and a pulling rate v<sub>T</sub>=0.033mm/s. Fig. 4 shows the flow field for different Marangoni numbers. In Fig. 4a (Ma=0) the velocities are comparable to the pulling rate except a small region,

close to the meniscus, where the velocities are vanishing. In this region, it is possible that the gas contained in the liquid or the micro-bubbles accumulate and are incorporated into the crystal when they attain a critical dimension. In such a situation, micro-bubbles are scarce and appear periodically. This has been observed by Wada [9] and Borodin [1]. It is shown in Fig. 4a to 4d that for increasing Marangoni numbers the flow field is radically modified and three significant aspects must be mentioned: a) the increase of velocity on the meniscus surface and in a region which has the same dimension that the meniscus height. The velocity has a value close to the pulling rate for small Marangoni numbers but increases dramatically for large Marangoni numbers. b) The velocities in the region where they were vanishing for Ma=0 increase and their values are comparable to the pulling rate. c) For a critical Marangoni number (~ 1.5) the apparition of a loop in the flow field is observed. This loop influences a region with a dimension comparable to the meniscus height. The apparition of this loop is accompanied by an increase of the velocities.



Fig. 4. Flow field for several Marangoni numbers.

The maximum velocity on the meniscus surface has a linear variation with the Marangoni number as can be seen in Fig. 5.



Fig. 5. Maximal velocity on the meniscus surface as a function of Marangoni number.

An increase of the thermal gradient establishes a linear increase of the maximum velocity on the meniscus surface but also an increase of the velocities in a region of the meniscus with dimensions comparable to the meniscus height. For the same Marangoni number (Ma) and for different Reynolds numbers in the meniscus (Re<sup>men</sup>) the flow field near the meniscus surface don't change significantly but the velocity values increase with the meniscus height (Fig. 6).

## 4. Conclusion

A lot of physical phenomena play an important role in the apparition of micro-bubbles in the shaped sapphire growth. The flow field in the capillary channel and in the meniscus has a major importance. The aim of this paper was to study numerically the flow field in the capillary channel as a function of the leading parameters (Re<sup>men</sup>, Re<sup>cap</sup>, Ma).

A strong influence of the Reynolds numbers in the meniscus (Re<sup>men</sup>) and in the capillary channel (Re<sup>cap</sup>), and also of the Marangoni number has been established.

The velocities in the liquid decrease when Re<sup>men</sup> increases and the maximum velocity position moves from the left of the exit of the capillary channel to the axis of symmetry. The maximum liquid velocity decreases when the Reynolds number in the meniscus increases in the same way for different Reynolds numbers in the capillary channel. It was found a critical value for Re<sup>men</sup> from which the maximum velocity remains constant. The maximum liquid velocity increases with the increase of the Reynolds number in the capillary channel.



Fig. 6. Flow field for Ma=8.670, Re<sup>cap</sup>=0.02 and various Re<sup>men</sup>.

The surface tension variation strongly influences the flow field. For a constant meniscus height (100 $\mu$ m) the flow field was studied for different Marangoni numbers. Till a critical value of Ma (~1.5) the liquid velocities are comparable to the pulling rate except a small region, close to the meniscus surface where the velocities are vanishing. For values of Ma greater than the critical value the apparition of a loop is observed, near the meniscus surface, with an increase of the liquid velocity. In this region near the meniscus surface, with dimensions comparable to the meniscus height, it is possible that the gas contained in the liquid or the micro-bubbles accumulate and are incorporated into the crystal when they attain a critical dimension. The maximum velocity of the meniscus surface has a linear variation with the Marangoni number. For a constant Marangoni number and for different Reynolds numbers in the meniscus, the velocities composing the loop increase with the increase of the meniscus height.

In order to build some explanations concerning the apparition and the distribution of microbubbles in shaped sapphire growth it is foreseen to perform numerical simulations with other geometries and to introduce the equation of conservation of the dissolved gas in the numerical model.

### References

- V. A. Borodin, T. A. Steriopolo, V. A. Tatarchenko, T. N. Yalovets, Izv. AN SSSR, Physics Series, 47, 368 (1983).
- [2] T. N. Yalovets, V. A. Borodin, A. E. Lezhnev, G. G. Sikhurulidze, T. A. Steriopolo, V. A. Tatarchenko, Izv. AN SSSR, Neorgan. Materialy, 21, 262 (1985).
- [3] I.E. Berezina, S.V. Tsivinskii, L. M. Zatulovskii, Izv. AN SSSR, Physics Series, 49, 2398 (1985).
- [4] V. A. Borodin, T. A. Steriopolo, V. A. Tatarchenko, T. N. Yalovets, Crystal Research and Technology, 20, 301 (1985).
- [5] T. Duffar, M. D. Serrano, L. Lerin, J. L. Santailler, Crystal Research and Technology, 34, 457 (1999).
- [6] F. Theodore, T. Duffar, J. L. Santailler, J. Pesenti, M. Keller, P. Dusserre, F. Louchet, V. Kurlov, Journal of Crystal Growth, 204, 317 (1999).
- [7] F. Theodore, Ph.D. Thesis, CEA-Grenoble 1997.
- [8] FIDAP, Version 8.0, User Manuals, Fluent Incorporated, 1998.
- [9] K. Wada, K. Hoshikawa, Journal of Crystal Growth, 50, 151 (1980)