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NUMERICAL SIMULATION OF HEAT TRANSFER IN TRANSPARENT AND SEMITRANSPARENT CRYSTAL GROWTH PROCESSES[1]

F. Barvinschi, T. Duffar[a], J. L. Santailler[b]

'Politehnica' Univ. of Timisoara, Dept. of Physics, P-ta Regina Maria, No. 1, 1900 Timisoara Romania
[a]Institut National Polytechnique de Grenoble, EPM, ENSHMG, BP 95, 38402 St Martin d´Heres, France
[b]CEA/CEREM/DEM, Commissariat a l´Energie Atomique, 17, rue des Martyrs, 38054 Grenoble, France

The finite element software FIDAP (FLUENT^{TM}) was used to calculate the thermal distribution in the sample and its surroundings during the Bridgman and Verneuil single crystal growth processes. All thermal exchanges present in the system are taken into account, including the internal radiation through semitransparent materials. The effects of transparency of BaF_2, CaF_2 and sapphire crystals is discussed and the major importance of the knowledge of the physical phenomena is discussed.

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1. Introduction

In the field of single crystals, there are materials whose optical properties are important, not only for their technological applications, but also during their melt growth processes. The theoretical study of semitransparent materials such as sapphire and fluoride crystals, is of great interest, despite the complexity of physical phenomena involved [1]. High quality crystal can be grown when the thermal environment surrounding the growing crystal is accurately controlled. However, the crystal growth process of a semitransparent material encounters the coupling of radiative, conductive and convective heat transport modes [2]. The numerical simulation of heat transport in complex crystal growth systems, such as Bridgman [3] and Verneuil [4] devices, is useful because it offers a precise knowledge of the growth process, enabling to tailor the optimum growth conditions. The control strategy for generating a desired temperature distribution during the crystal growth process is mainly limited by our understanding of the physical phenomena. Considering the high melting temperatures (1628 K for BaF_2, 2323 K for sapphire) and thanks to the evidence of experimental results [5], it has been anticipated that the incorporation of the participating media radiation in the modelling of such crystal growth processes could have a major effect on the accuracy of the analysis [6].

This work is a review of the results of several research programs on the Bridgman method, used for fluoride crystal growth [6, 8], and on the Verneuil method, dedicated to sapphire growth [4]. The finite element software FIDAP was used to simulate, under some assumptions, the heat transport phenomena involved in the Bridgman and in the Verneuil crystal growth methods. All the thermal

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exchanges into the set-up are taken into account, including the wall-to-wall radiation between surfaces separated by a fully transparent or by a semitransparent medium.

The case of semitransparent material, such as fluoride samples which absorb and emit radiation both in the solid and in the liquid phases, has been studied. A comparison between an opaque, a fully transparent and a semitransparent material is performed and the effects of some parameters, such as the optical absorption coefficient and thermal conductivity, on the solid-liquid interface shape and on the thermal gradients are discussed. Two physical models developed for internal radiation phenomena, i.e. the Rosseland diffusion approximation [3] and the P-1 method [7], have been used. We will focus on the numerical limitations associated to these models used in conjunction with the wall-to-wall radiation model.

2. Vertical Bridgman method for growing fluoride crystals

In the Vertical Bridgman (VB) method, the material, confined into cylindrical crucible, is molten and slowly re-solidified by translating the crucible through the furnace. It is known that the shape of the solid-liquid interface, which is determined by the thermal fluxes, has a strong influence of the crystal quality. Therefore, it is of importance to accurately control the thermal field during the solidification process. The VB process is the most common method of growing fluoride crystals, such as BaF_2 and CaF_2, which emit and absorb radiation both in the solid and in the liquid phases, thus being semitransparent materials. In fact, the optical absorption coefficient of the molten phase is higher than that of the solid phase, but not so high as to consider the fluoride liquid as opaque [5].

The VB process has been modelled by using the finite element software FIDAP [7], employing realistic assumptions in order to simulate the growth of BaF_2 and CaF_2 samples. The mathematical formulation of the small-scale model (see Fig. 1), the operating parameters, as well as the physical properties of the system are detailed in [8, 9]. In this axi-symmetric model, the crucible wall exchanges heat by conductive and radiative transfer with an idealised axial furnace temperature profile $T_f = f(y)$.

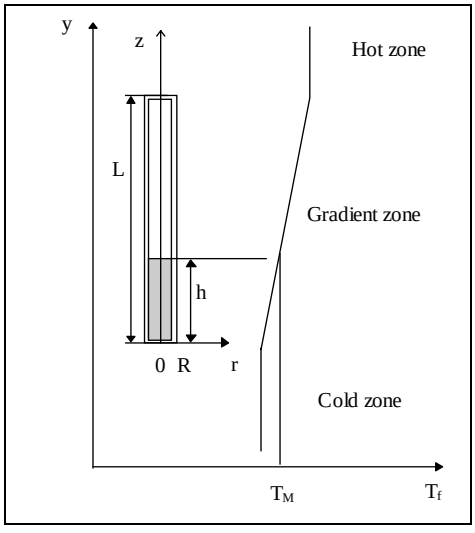


Fig. 1. Mathematical description of the Vertical Bridgman system for growth of semitransparent crystals.

In order to take into account the heat transport by radiation in the crystal and in the melt, two approximations were used: (i) the Rosseland diffusion approximation [10] for rather opaque materials and (ii) the P-1 approximation [7] for rather transparent materials.

(i) In the Rosseland approximation the medium is considered as a solid with a temperature dependent thermal conductivity:

$$k = k^{\text{mol}} + \frac{16n^2\sigma T^3}{3a} \tag{1}$$

where k^{mol} is the molecular conductivity, n is the refractive index of the medium, σ is the Stefan-Boltzmann constant and a is the Rosseland mean optical absorption coefficient.

The temperature distribution in the melt and in the crystal is determined by considering the thermal conductivity (1) in the steady-state energy balance:

$$\nabla\left(k\nabla T\right) = 0 \tag{2}$$

The reference case where the melt and the crystal are both opaque is also solved ($k = k^{mol}$).

(ii) In order to simulate the additional mechanism of internal radiation in a more reliable way, the P-1 approximation [7] has also been used. In this method the coupled conduction-radiation transport through an optically thick material, with grey behaviour, is considered by solving two equations, accounting for radiative heat flux in a participating medium:

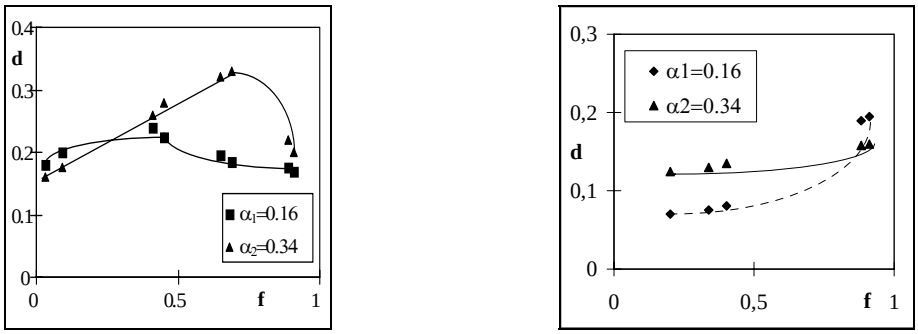
$$\nabla\left(k\nabla T\right) + \frac{1}{3a}\Delta\bar{J} = 0 \tag{3}$$

$$\Delta\bar{J} = 3a^2\left(\bar{J} - 4n^2\sigma T^4\right) \tag{4}$$

where $\bar{J}$ is the mean irradiance, $\frac{1}{3a}\Delta\bar{J}$ acting as an additional volumetric heat source due to radiation and being used in the energy equation. Thus, the conservation equation of energy (3) is evaluated in conjunction with the approximate form (4) of the complex equation of the radiation intensity through an absorbing and emitting medium [10]. The medium is assumed to be grey, i.e. there is no dependence on wavelength.

Unfortunately, the FIDAP software does not allow the use of the participating media radiation option simultaneously with the wall-to-wall radiation capability. Despite this drawback, the P-1 model has been shown to predict some proper trends in the case of optically tick materials, such as silica. The results obtained in the P-1 approximation were compared with those resulted when the material was assumed as fully transparent (a = 0) or opaque (a = ∞).

We analyse now the results obtained in the Rosseland approximation from several steady-state heat transport calculations, by using the aspect ratio, $\alpha = D/L$, as parameter ($D = 2R$ and L are the crystal diameter and the sample length, respectively). The solidified fraction, f, is defined as the ratio between the grown crystal length, h, at various stages of the growth process and the sample length, L, while the non-dimensional curvature of the liquid-solid interface, d, is the ratio between the interface deflection, $\delta = h(0)-h(R)$, and the crystal radius, R. In Fig. 2 the dimensionless interface deflection is shown as a function of the solidified fraction, for two aspect ratio values, corresponding to the BaF_2 samples of α_1= 11 mm/70 mm and α_2= 24 mm/70 mm.



a) b)

Fig. 2. The non-dimensional curvature of the liquid-solid interface, d, as a function of the solidified fraction, f, for two aspect ratio values, in the case of an opaque (a) and a semitransparent (b) BaF_2 sample.

The curvature of the liquid/solid interface depends on the solidified fraction and on the aspect ratio, for both opaque and semitransparent samples: the liquid-solid interface deflection is smaller for a small aspect ratio than for a larger one, as well as for a semitransparent material than for an opaque one. As the charge is semitransparent, the axial heat transfer is increased by radiation and this leads to a higher axial flux than in the pure opaque case. Therefore, the semitransparent crystal experiences a continuous increase of the liquid/solid interface deflection during the solidification process.

Typical thermal distributions obtained when the P-1 approximation is used are shown in Fig. 3, where T_M is the melting isotherm. The liquid-solid interface position shifts downward, toward the cold zone, as the charge becomes more opaque to radiation. It is important to note that we consider a silica crucible. Thus, a_a represents the optical absorption coefficient of the silica, while a_s denotes the optical absorption coefficient of the CaF_2.

The liquid-solid interface deflection increases as the optical absorption coefficient increases, the transparent CaF_2 charge showing the most flatter solidification isotherm. This is in qualitative agreement with the results obtained in [1-3] and with the experimental results of [11].

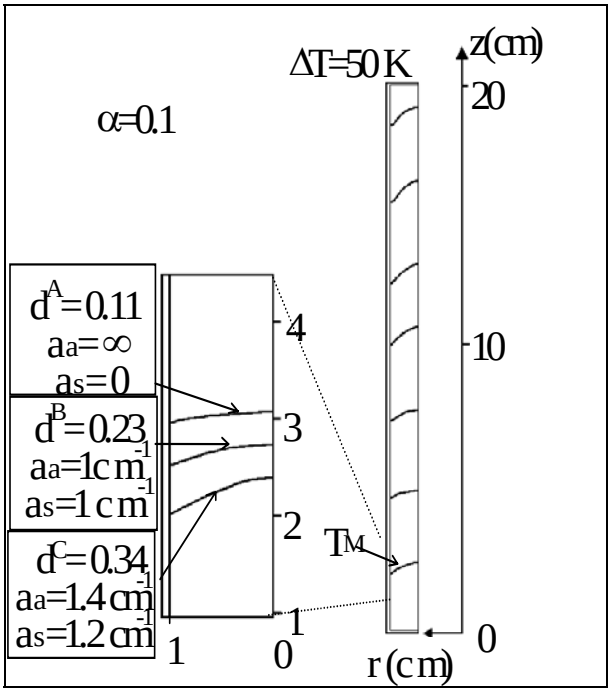


Fig. 3. Crystallisation interface locations and shapes for different values of the optical absorption coefficient, calculated in the P-1 approximation; A: $a_a = \infty$, $a_s = 0$; B: $a_a = 1$ cm^{-1}, $a_s = 1$ cm^{-1}; C: $a_a = 1.4$ cm^{-1}, $a_s = 1.2$ cm^{-1}.

3. Verneuil method for growing sapphire crystals

In the Verneuil sapphire crystal growth system, the alumina powder is molten during its fall into an oxygen-hydrogen flame. The liquid droplets fall on the top of the slowly-pulled crystal. From a physical point of view, the Verneuil process is complex because of the coupling of the turbulent combustion between hydrogen and oxygen, the hydrodynamics of the gas phase and the heat transport by convection, conduction and radiation.

Numerical simulations of the Verneuil crystal growth process have been carried out using the FIDAP finite element software, and the results have been presented in a previous paper [4]. In order to determine the effect of the transparency of the sapphire, some assumptions were used: the process is considered in quasi-steady state, the model is axi-symmetric and the latent heat of fusion is negligible, i.e. the powder is not included in the model. The results obtained in the case where the crystal is considered as opaque have been compared with the simulation of a fully transparent material.

The transparent crystal appears to be cooler than the opaque one, because in a transparent body the absorption of heat is reduced. The axial thermal gradient in the opaque crystal is smaller than that calculated in the case of a transparent crystal. By comparing the numerical results with experimental thermocouple data, it has been concluded that the simulation of a fully transparent sapphire crystal is more realistic than that of an opaque one.

A further step in this study will be to analyse the effect of the participating media radiation through the sapphire.

4. Conclusions

The Bridgman and Verneuil methods are used for the production of crystals of a wide range of materials, owing to their simplicity and cheapness. From the point of view of physical phenomena, the solidification of transparent and semitransparent materials involves the coupling of heat exchanges by radiation, conduction and convection.

The finite element software FIDAP was used to simulate, under such assumptions, the effect of the transparency on the solidification process in the case of fluoride crystals (BaF_2, CaF_2) obtained by the vertical Bridgman method. From these calculations, it is found that the liquid-solid interface curvature depends on the degree of transparency. The liquid-solid interface deflection is higher for opaque crystals than that for semitransparent ones, in the Rosseland diffusion approximation. In the limits of the P-1 approximation, the liquid-solid interface deflection increases as the optical absorption coefficient increases, the transparent charge showing the most flat solidification isotherm. Results obtained with both models are in qualitative agreement and are also in agreement with some experimental observations that confirm that the effect of transparency of both the molten and solid phases is of importance in the growth of such materials.

Looking at the steady-state numerical model of the Verneuil growth process, it is found that the more realistic results, close to the experimental data, are obtained when the sapphire is considered as a fully transparent material. Provided that all major physical phenomena are well implemented and some empirical parameters of the model are adjusted, the numerical results are in qualitative agreement with experimental observations.

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