Some properties of the stationary solution in the case of solidification using Bridgman technique

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Abstract: - A stationary free boundary model of solidification in the case of the vertical Bridgman crystal growth technique is considered. The nonlinear boundary problem of the Navier-Stokes and heat equations is solved numerically in an axi-symmetric domain using FreeFem++ software. Also, using qualitative studies some theoretical properties of the solution of nonlinear boundary problem are obtained.

Key-Words: - Vertical Bridgman, Stationary problem, Free boundary problem

1 Introduction
In 1924, Bridgman developed a method for growing crystals in a cylindrical crucible. The crystal grows as the crucible moves in a certain temperature configuration and layout of the furnace [1]. Most known temperature configurations are Grenoble (corresponding to adiabatic insulation among the ampoule’s sidewall) and MIT (corresponding to three zones of temperature among the ampoule’s sidewall). As for the layout of the furnace, it can be vertical or horizontal.

The Bridgman technique is used to grow single crystals of As, Ag, LiI, SiAs, GaAs, GaGe, etc. In the case of binary alloys, the rejection of the dopant at the solidification interface represents a serious problem for practical crystal growers. Hence, the properties of the semiconductor crystals are strongly dependent of the dopant rejection which is controlled by the shape of the solid/liquid interface. On the other hand, the interface is a free boundary, unknown a priori, reason for which this kind of problems request more theoretical investigations. In literature, there are some investigations based on the model proposed in [2], but they were made under the assumption that the solidification interface is a priori known [3-7].

In this paper the free boundary model proposed by Chang and Brown is considered [2]. The furnace configuration is of MIT type, i.e., the furnace presents three zones: (i) the hot zone; (ii) the gradient zone; (iii) and the cold zone.

Some properties of the solution for the considered boundary value problem are established. These are validated through numerical simulations, based on a fixed point algorithm, performed using FreeFem++ software.

2 Problem Statement
Let \( \Omega_i \) be the domain occupied by the melt, and \( \Omega_s \) the domain occupied by the crystal (\( \Omega = \Omega_i \cup \Omega_s \)):

\[
\Omega_i = \{ (r,z) \in \mathbb{R}^2 | 0 \leq r < R \text{ and } 0 < z < h(r) \}
\]

\[
\Omega_s = \{ (r,z) \in \mathbb{R}^2 | 0 \leq r < R \text{ and } h(r) < z < A \}
\]

where \( h(r) \) is the function describing the solidification interface satisfying \( h(R) = \frac{A}{2} \). A schematic representation of the domains is given in Fig. 1.

![Fig. 1: The domains occupied by melt and solid, the corresponding boundaries, and the temperature profile in the furnace.](image-url)
According to [2], the dimensionless form of the Navier-Stokes and heat equations in the liquid and crystal is given by:

\[
\begin{align*}
\overline{\nabla} \overline{u} &= 0 \text{ in } \Omega_l \\
(\overline{\nabla} \overline{\nabla}) \overline{u} &= -\overline{\nabla} p + \overline{P} r \Delta \overline{u} + \overline{R} r \overline{\theta} \overline{e}_z \text{ in } \Omega_l \\
\overline{\nabla} \overline{\nabla} \theta &= \Delta \theta \text{ in } \Omega_l \\
\overline{u}_c \cdot \overline{e}_z &= -P e \overline{e}_z \text{ in } \Omega_s \\
\overline{u}_c \overline{\nabla} \theta_c &= \gamma \Delta \theta_c \text{ in } \Omega_s
\end{align*}
\]

where \( \overline{u} \) represents velocity field in the melt, \( \theta \) - the temperature in the melt, \( \overline{u}_c \) - translation rate, and \( \theta_c \) - temperature in the crystal. The dimensionless equations are obtained by scaling the dimensional parameters as follows:

\[
\begin{align*}
\overline{u} &= \frac{L}{\alpha_i} \overline{u}^* \quad p = \frac{L^2}{\rho_i \alpha_i} \overline{p}^* \quad \overline{x} = \frac{x^*}{L} \quad T = \frac{T^* - T_s}{T_c - T_s} \\
\end{align*}
\]

where \( L \) represents the length of the ampoule, \( \alpha_i \) - the thermal diffusivity in melt, \( \rho_i \) - the density of melt, \( T_c \) - temperature of the cold zone, \( T_s \) - temperature of the hot zone. Following computations in dimensionless form, the new parameters are introduced: Rayleigh number \( Ra = \frac{\beta_i g (T_c - T_s) L^3}{\alpha_i \nu} \) (\( \beta_i \) is the thermal expansion coefficient, \( g \) - the gravitational constant, \( \nu \) - the kinematic viscosity of melt), Prandtl number \( Pr = \frac{\nu}{\alpha_i} \), Péclet number \( Pe = \frac{V_s L}{\alpha_i} \) (\( V_s \) - growth rate), and \( \gamma \) - ratio of the solid and melt thermal diffusivities.

The boundary conditions corresponding to problem (1) are:

\[
\overline{u} \mid_{\Gamma_1}, \overline{u}_c \mid_{\Gamma_2}, \overline{u}_c \mid_{\Gamma_3} = \overline{u}_r \quad (2) \\
\overline{u}_c \mid_{\Gamma_1}, \overline{u}_c \mid_{\Gamma_2} = \overline{u}_r \quad (3) \\
\overline{u} \cdot \overline{t} \mid_{\Gamma_5} = Pe \cdot \overline{t} \\
\sigma(\overline{u} \cdot \overline{n}) \mid_{\Gamma_5} = Pe \cdot \overline{n} \\
\overline{\theta} \mid_{\Gamma_1} = 0 \\
\overline{\theta \mid_{\Gamma_2}} = \frac{1}{L_c} z + \frac{L_g - A}{2 L_c} \quad , \quad z \in \left[ \frac{A}{2} - \frac{L_g}{2} \right] \\
\overline{\theta \mid_{\Gamma_3}} = 1 \quad , \quad z \in \left[ \frac{A}{2} + \frac{L_g}{2} \right] \\
\overline{\theta \mid_{\Gamma_4}} = \frac{1}{L_g} z \quad , \quad z \in \left[ \frac{A}{2} - \frac{L_g}{2} \right] (7)
\]

where \( \overline{u}_r = u_r \cdot \overline{e}_z \) is the dimensionless velocity of translation, \( \overline{t} \) - tangent unit vector to the solidification interface, \( \overline{n} \) - outward normal unit vector, \( \sigma \) - ratio of the solid and melt densities, \( A \) - dimensionless length of the ampoule, \( L_g \) - dimensionless length of gradient zone, \( k \) - ratio of the solid and melt thermal conductivities, \( S = \frac{\Delta H_l}{\rho c_p(T_c - T_s)} \) - dimensionless Stefan number (\( \Delta H_l \) is the latent heat of melt, \( c_p \) - heat capacity of melt).

The translation of the ampoule in the furnace is simulated by supplying melt into the ampoule at \( z = A \), and pulling crystal at \( z = 0 \). The sidewall of the ampoule is assumed to be a no slip surface (conditions (2)-(3)). Conditions (4)-(5) refer to no slip tangential to the crystal and a solidification rate proportional to the growth rate. Conditions (6)-(8) express the imposed temperature value on the ampoule’s walls. Condition (9) refers to a constant temperature at the solidification interface, and condition (10) to a prescribed heat flux at the solidification interface.

3 Some Properties of BVP’s solution

In order to determine the solution’s properties for problem (1)-(10), the boundary conditions for the Navier-Stokes equation are first homogenized. After that, a global equation for the temperature field is considered (based on the heat equations in melt, solid and the corresponding boundary conditions).

Let \( (\overline{u}, \overline{u}_c, \overline{\theta}, \overline{\theta}_c) \) be a solution of the problem (1)-(10), \( \overline{u}_i = \overline{u} - \overline{u}_r \) and \( \overline{u}_i = \overline{u}_c - \overline{u}_r \) the velocity fields obtained after homogenizing the Navier-Stokes’ boundary conditions. Denoting by

\[
\Theta(r, z) = \begin{cases} 
\overline{\theta}, & \text{for } (r, z) \in \Omega_i \\
\overline{\theta}, & \text{for } (r, z) \in \Omega_s 
\end{cases}
\]

the “global temperature”, and defining the coefficients \( \bar{\alpha} \) and \( \beta \) as follows:

\[
\begin{align*}
\theta \mid_{\Gamma_1} &= 1 \\
\theta \mid_{\Gamma_3} &= \frac{T_c - T_s}{T_c - T_r} = 0.5 \\
\left[ (\overline{u} \cdot \overline{n}) \mid_{\Gamma_5} - k (\overline{u} \cdot \overline{n}) \right] \mid_{\Gamma_5} &= S Pe n_z \\
\end{align*}
\]
\[ \alpha(r, z) = \begin{cases} \bar{u}, & \text{for } (r, z) \in \Omega_i \\ 0, & \text{for } (r, z) \in \Omega_s \end{cases} \]

\[ \beta(r, z) = \begin{cases} 1, & \text{for } (r, z) \in \Omega_i \\ \gamma, & \text{for } (r, z) \in \Omega_s \end{cases} \]

the problem (1) becomes:

\[ \nabla \bar{u}_1 = 0 \text{ in } \Omega_i \]

\[ (\bar{u}_1 \nabla) \bar{u}_1 = -\nabla p + Pr \Delta \bar{u}_1 + Ra Pr \Theta |_{\Omega_i} \bar{e}_z - \]

\[ - (\bar{u}_1 \nabla) \bar{u}_1 - (\bar{u}_1 \nabla) \bar{u}_1 - (\bar{u}_1 \nabla) \bar{u}_1 \text{ in } \Omega_i \]

\[ \alpha \nabla \Theta = \beta \Delta \Theta - \bar{u}_\gamma \nabla \Theta \text{ in } \Omega \]

\[ \bar{u}_{1c} = 0 \text{ in } \Omega_s \]

Note that \( \Theta, \bar{u}, \alpha \) and \( \beta \) are continuous functions defined on \( \Omega \). The corresponding boundary conditions are:

\[ \bar{u}_1 |_{r_2, r_3} = 0 \]

\[ \bar{u}_{1c} |_{r_1, r_2} = 0 \]

\[ \bar{u}_1 \cdot \bar{e}_z = (Pe - u_{\gamma}) \cdot \bar{e}_z = 0 \]

\[ \sigma(\bar{u}_1 - \bar{u}) |_{r_3} = (Pe - \sigma u_{\gamma}) \cdot n_z = 0 \]

\[ \Theta |_{r_1} = 0 \]

\[ \Theta |_{r_2} = r \]

\[ \Theta |_{r_3} = 1 \]

\[ \Theta |_{r_3} = \frac{T_c - T_r}{T_c - T_r} = 0.5 \]

\[ \{ (\bar{u}_1 \nabla \Theta), -k(\bar{u}_1 \nabla \Theta) \} |_{r_3} = Sp e n_z \]

Multiplying Eq.

\[ \alpha \nabla \Theta = \beta \Delta \Theta - \bar{u}_\gamma \nabla \Theta \]

by \( \Theta \) and integrating over \( \Omega \), we get:

\[ \int_{\Omega_i} \alpha \nabla \Theta \cdot \Theta = \int_{\Omega_i} \beta \Delta \Theta \cdot \Theta - \int_{\Omega_i} \bar{u}_\gamma \nabla \Theta \cdot \Theta. \]

Because

\[ \int_{\Omega_i} \alpha \nabla \Theta \cdot \Theta = 0, \]

\[ \int_{\Omega_i} \beta \Delta \Theta \cdot \Theta = -\beta \int_{\Omega} |\nabla \Theta|^2 + \frac{1}{2} \beta SPe \int_{r_3} n_z, \]

\[ \int_{\Omega_i} \bar{u}_\gamma \nabla \Theta \cdot \Theta = \frac{R u_{\gamma}}{2} + \frac{1}{8} u_{\gamma} \int_{r_3} n_z, \]

we obtain the inequality

\[ \int_{\Omega} |\nabla \Theta|^2 \leq \frac{1}{\beta} \left[ \frac{R u_{\gamma}}{2} + \frac{1}{8} u_{\gamma} \int_{r_3} n_z + \frac{1}{2} \beta SPe \int_{r_3} n_z \right] \leq \frac{1}{\beta} \left[ \frac{R u_{\gamma}}{2} + \frac{4 \beta SPe + u_{\gamma}}{8} \right] \]

or

\[ \| \nabla \Theta \|_2 \leq \frac{1}{\beta} \left[ \frac{R u_{\gamma}}{2} + \frac{4 \beta SPe + u_{\gamma}}{8} \right] (15) \]

Relation (22) implies that the growth of the temperature field is finite in \( \Omega \). Since \( \Theta |_{r_1} = 0 \), it follows that \( \Theta \) is finite, and hence \( \| \Theta \|_{L^2} \leq C_1 \), with \( C_1 \) a real positive constant.

Let \( X_0 = \{ \bar{u} \in (H^1(\Omega_i)) | \nabla \bar{u} = 0 \} \). Multiplying Eq.

\[ (\bar{u}_1 \nabla) \bar{u}_1 = -\nabla p + Pr \Delta \bar{u}_1 + Ra Pr \Theta |_{\Omega_i} \bar{e}_z - \]

\[ - (\bar{u}_1 \nabla) \bar{u}_1 - (\bar{u}_1 \nabla) \bar{u}_1 - (\bar{u}_1 \nabla) \bar{u}_1 \]

by \( \bar{u}_i \in X_0 \), and integrating over \( \Omega_i \), we get:

\[ \int_{\Omega_i} (\bar{u}_1 \nabla) \bar{u}_1 = -\int_{\Omega_i} \nabla p \bar{u}_1 + \int_{\Omega_i} Pr \Delta \bar{u}_i \bar{u}_1 + \]

\[ + Ra Pr \int_{\Omega_i} \Theta |_{\Omega_i} \bar{e}_z \bar{u}_i - \int_{\Omega_i} (\bar{u}_1 \nabla) \bar{u}_1 \bar{u}_i - \]

\[ - \int_{\Omega_i} (\bar{u}_1 \nabla) \bar{u}_i \bar{u}_i - \int_{\Omega_i} (\bar{u}_1 \nabla) \bar{u}_i \bar{u}_i \]

or

\[ Ra Pr \int_{\Omega_i} \Delta \bar{u}_i \bar{u}_i + Ra Pr \int_{\Omega_i} \Theta |_{\Omega_i} \bar{e}_z \bar{u}_i = 0. \]

This leads to

\[ \| \nabla \bar{u}_i \|_{L^2} \leq \int_{\Omega_i} | \nabla \bar{u}_i |^2 = Ra \int_{\Omega_i} \Theta |_{\Omega_i} \bar{e}_z \bar{u}_i \leq Ra \| \Theta \|_{L^2} \| \bar{u}_i \|_{L^2}. \]

Applying the Friedrighs inequality, which in our case is written

\[ \| \bar{F} \|_{L^2} \leq \sqrt{A^2 + R^2} \| \nabla \bar{u}_i \|_{L^2} \]

we get

\[ \| \nabla \bar{u}_i \|_{L^2} \leq \sqrt{A^2 + R^2} Ra C_1 < \infty. \]

Inequality (24) implies that the growth of the velocity field is finite in \( \Omega \). Since \( \bar{u}_i |_{r_1} = 0 \), it follows that \( \bar{u}_i \) is finite, or \( \bar{u} = \bar{u}_i + \bar{u}_\gamma \) is finite.

4 Numerical Example

In the following, numerical simulations, based on a fixed point algorithm, are performed using FreeFem++ software [8]. The values for the parameters involved in (1)-(10), corresponding to Ga-doped Ge grown in terrestrial gravity conditions (\( Ra = 10^6 \)), are given in Table 1.
Table 1: Values of the parameters

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>1</td>
<td>$\mu_r$</td>
<td>0.01</td>
</tr>
<tr>
<td>$L_g$</td>
<td>0.125</td>
<td>$\gamma$</td>
<td>1</td>
</tr>
<tr>
<td>$Pr$</td>
<td>0.01</td>
<td>$k$</td>
<td>1</td>
</tr>
<tr>
<td>$Ra$</td>
<td>$10^6$</td>
<td>$S$</td>
<td>1</td>
</tr>
<tr>
<td>$Pe$</td>
<td>0.01</td>
<td>$\sigma$</td>
<td>1</td>
</tr>
</tbody>
</table>

The algorithm for solving problem (1)-(10) takes as input data the functions $h^{(0)}(r) = \frac{A}{2}$, $\bar{u}^{(0)}(r, z) = \bar{u}_r$ and $\Theta^{(0)}(r, z) = \tau$, and computes the unknown functions $h(r)$, $\bar{u}(r, z)$ and $\Theta(r, z)$. For computing numerical solution, the following steps are used:

1. The “global” heat equation with the boundary condition (20) is solved.
2. The isotherm corresponding to condition (19) is found.
3. The domain’s deformation is constructed, in order to overlap the boundary to the isotherm found at step 2.
4. The Navier-Stokes equation on de deformed domain is solved.
5. The steps 1-4 are repeated until the both variations of temperature field and velocity field become less than a sufficiently small error, $\varepsilon$.

A schematic representation of the domain deformation is given in Fig. 2.

![Fig. 2: The domain deformation](image)

The computed temperature and velocity fields are presented in Fig. 3.

![Fig. 3: Temperature and flow field in the stationary case for Ga-doped Ge grown in terrestrial conditions](image)

The behaviour of the flow field in the melt is determined by the Rayleigh number $Ra = 10^6$, corresponding to the terrestrial conditions. Two convection cells are observed: a strong one, in the lower part of the melt, and a weaker one in the upper part. The computed streamlines are equally spaced between the maximum (0.1526) and the minimum (-0.0118) values.

For the temperature field, it can be observed that the shape of the isotherms tend to flatten in melt toward the solidification interface. Because the velocity in crystal is constant, and equal to the growth rate, the shape of the isotherms in crystal is not affected by the gravity condition. The computed isovalues for temperature are equidistant between a minimum (0) and a maximum (1) value.

A larger view of the region containing the computed solidification interface is presented in Fig. 4.

![Fig. 4: The solidification interface for $Ra = 10^6$.](image)

It should be underlined that in previous investigations [3-7], the numerical solution of the problem (1)-(10) was computed under the assumption that the solidification interface is a straight line, on which a constant temperature is imposed, but the heat flux at the interface is neglected. The approach proposed here is more realistic because it considers both conditions of constant temperature and prescribed heat flux at the interface.
4 Conclusions
A free boundary stationary model for the Bridgman crystal growth technique was considered. From the properties of the considered BVP’s solution, upper limitations for the velocity field and temperature are estimated. Further investigations of the present work will focus on the non-stationary case able to give more useful information for optimizing crystal growth processes.

References: