CRYSTAL GROWTH INTERFACE

AND

MICROGRAVITY

Master Thesis

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Màster en Enginyeria Química i de Processos

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# Nomenclature

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<th>Definition</th>
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<td>2D</td>
<td>Two dimensions</td>
</tr>
<tr>
<td>3D</td>
<td>Three dimensions</td>
</tr>
<tr>
<td>$a$</td>
<td>Steepness of temperature gradient</td>
</tr>
<tr>
<td>$A_{mash}$</td>
<td>Mushy constant parameter</td>
</tr>
<tr>
<td>BN</td>
<td>Boron Nitride</td>
</tr>
<tr>
<td>CFD</td>
<td>Computation Fluid Dynamics</td>
</tr>
<tr>
<td>$c_p$</td>
<td>Heat capacity (J/K·kg)</td>
</tr>
<tr>
<td>$dh$</td>
<td>Solid/liquid interface</td>
</tr>
<tr>
<td>$f$</td>
<td>Phase change temperature function</td>
</tr>
<tr>
<td>$g$</td>
<td>Gravity (m/s$^2$)</td>
</tr>
<tr>
<td>GaSb</td>
<td>Gallium Arsenide</td>
</tr>
<tr>
<td>$Gr$</td>
<td>Grashof Number</td>
</tr>
<tr>
<td>$H$</td>
<td>Enthalpy (J/K·kg)</td>
</tr>
<tr>
<td>$h_{sens}$</td>
<td>Sensible enthalpy (J/K·kg)</td>
</tr>
<tr>
<td>$h_{ref}$</td>
<td>Reference enthalpy (J/K·kg)</td>
</tr>
<tr>
<td>$htan$</td>
<td>Hyperbolic tangent</td>
</tr>
<tr>
<td>$h_c$</td>
<td>Solid height on the centre of the crucible (m)</td>
</tr>
<tr>
<td>$h_s$</td>
<td>Solid height in the sides of the melt/crystal (m)</td>
</tr>
<tr>
<td>$k$</td>
<td>Thermal conductivity (W/K·m$^2$)</td>
</tr>
<tr>
<td>$L$</td>
<td>Crucible longitude in {0,1,0} direction (m)</td>
</tr>
<tr>
<td>$La$</td>
<td>Total latent heat (J/kg)</td>
</tr>
<tr>
<td>$Num$</td>
<td>Number of time steps</td>
</tr>
</tbody>
</table>
\( p \)  
Drop temperature rate parameter (K/s)

\( PR \)  
Pulling rate of the crucible (m/s)

\( Pr \)  
Prandtl Number

\( PR_c \)  
Growth velocity from the centre of the interface

\( R_i \)  
Melt/crystal radius (m)

\( R_a \)  
Ampoule total radius (m)

\( Re \)  
Reynolds number

\( S \)  
Source term

\( T \)  
Temperature (K)

\( T_c \)  
Hot/ top temperature (K)

\( T_h \)  
Cold/bottom temperature (K)

\( T_{lin} \)  
Linear temperature along lateral walls distribution (K)

\( T_{htan} \)  
Hyperbolic tangent temperature distribution along lateral ampoule walls (K)

\( T_{pc} \)  
Phase change temperature (K)

\( t \)  
Current time (s)

\( u \)  
Fluid velocity (m/s)

\( VB \)  
Vertical Bridgman

\( y \)  
Each node value of the \{0,1,0\} array with L longitude (m)

\( \gamma_{pc} \)  
Interface initial position in \{0,1,0\} direction (m)
### Greek Symbols

- $\alpha$: Thermal diffusivity ($\text{m}^2/\text{s}$)
- $\beta$: Liquid fraction
- $\beta_T$: Thermal expansion coefficient ($\text{K}^{-1}$)
- $\Delta H$: Latent heat ($\text{J/kg}$)
- $\Delta T_s$: Total effective gradient temperature ($\text{K}$)
- $\Delta t$: Time step size ($\text{s}$)
- $\mu$: Dynamic viscosity ($\text{kg/m}\cdot\text{s}$)
- $\rho$: Density ($\text{kg/m}^3$)
- $\nu$: Kinematic viscosity ($\text{m}^2/\text{s}$)
1. Introduction

The material research has become a critical spot on modern society. Nowadays, there is a notable link between the new upcoming materials and the improvement of many modern devices. There is a lot of research dedicated on crystal growth and dopant distribution on semiconductors, which are responsible of the modern electronic industry [1].

A hindrance that comes up on the development and production of semiconducting materials is the convective currents that occur during the directional solidification. The currents near to the solid-liquid interface of the system provoke instabilities on the crystal surface, leading to a not flat interface shape and to a not homogeneous distribution of the dopant in it. Those effects are directly associated to the buoyancy effect when there is a temperature gradient in the system in Earth laboratories.

The concept of semiconductors in microgravity opens in the seventies a huge spectrum of new possibilities of quality improvement for these materials as convective currents can be greatly diminished. The potential of the microgravity concept has, therefore, brought an interest on having a complete understanding on the interface behaviour of crystal growth encapsulated systems. Notwithstanding, this investigation front has not been exploited yet because of the problems related with the impact of the residual accelerations inherent to the satellite motion (for instance g-jitter effects) on the quality of the crystal grown under these conditions.

The table 1 shows the major considered hindrances that occur during the directional growth.

There is evidence that the gravity role is an important factor to take into account. The most important gravity dependant factor resides on the melt, which are the buoyancy convections, as mentioned previously. However, other mechanisms not directly associated with the gravity role as forced convection, interface kinetics, need to be still regarded.
This fact leads to consider the potential of doing a pioneer and useful research fully dedicated on this theme, focusing the target on performing Computational Fluid Dynamic (CFD) simulations of a Bridgman system in microgravity. This preliminary study would be enough to assess the interface behaviour and identify the occurring phenomena.

The fact of performing this type of simulations is also a matter of capital importance due to the huge economical and technological difficulties of performing such experiments on the space. It is impossible to create a realistic system under microgravity on the earth, considering that, in some cases, a Bridgman growth requires days to be completed. Parabolic aeroplanes, for instance only reach microgravity around 30 seconds.

The Bridgman system is a technique used for the directional growth of basically bulk crystals of semiconductors. It is composed by an ampoule that contains the melt semiconductor, and a
solid part of it called seed. The semiconductor that will be studied is GaSb and the ampoule is composed of Boron Nitride (BN). This method is usually used both in horizontal and vertical orientations, depending on the semiconductor.

![Figure 1 Vertical Bridgman (VB) scheme with the T distribution on the wall of the crucible](image)

The ampoule is introduced into the furnace so that the seed is not above the melting temperature of the semiconductor. By this point, the ampoule is dragged out the furnace progressively in a constant ambient temperature until the semiconductor is totally solidified.

Typically CFD simulations of Bridgman systems are usually performed to gain knowledge of the heat, flow and dopant distribution under magnetic fields [2,6,7,9,10] and also under the accelerated crucible rotation [2-5]. Also, many specialized papers concerns Bridgman growth under microgravity conditions [4,7,8].

**Hypothesis**

This project is focused on having knowledge about the solid-liquid interface behavior of confined semiconducting materials while performing directional crystal growth. A first preliminary simulation of a melting exercise will provide the validity of the following simulation [11]. The main objective is to perform satisfactory computational fluid dynamic simulations under low gravity conditions.
The next step is to determine the interface deflection through time and quantify this deformation. The main purpose of this study is to confirm that low gravity affects to the convective currents so that no significant deflection occurs during crystallization. The expected results are that interface deformation will not be significant.

By confirming this phenomenon, this project will provide a guaranty of validation on those simulators that assume a priori flat interfaces in their calculations. There is a preference on using this type of flat interface simulators such as COMGA (Institute of Advanced Problems in Mechanics, RAS) rather than conventional ones due to their fast performance, so a validity to operate under these conditions is required.

2. Model description and numerical method

Assumptions

Before doing the main paper simulation, a basic approach [11] on the melt interface behaviour has been performed in FLUENT© as a calibration and validation point of some intrinsic calculation parameters.

Considering this project as a master thesis, the main approach will regard the following limitations:

- The melt will be constituted of pure GaSb, no dopants are considered
- The ampoule will only be considered in the lateral walls. So the top and bottom of the ampoule will lack of thickness, although virtual wall of the ampoule will be considered for the simulation consistency.
- Radiation, wall resistance effects and magnetic fields are not considered.
- There is no seed to start the crystallization.

The changing parameters that will be considered for this approach are:

- Gravity orientation: vertical, diagonal (45º) and horizontal.
- Wall temperature: linear and hyperbolic tangent distribution.
- 2D and 3D meshes contrast.
System description

The figures 2(a) and 2(b) illustrate the Bridgman considered systems in 2D and 3D in Cartesian coordinate (x,y,z).

All the cases are solved in transient conditions and they are considered non-axysimmetric due to the different gravity orientations. The discretization of the momentum equations are truncated on the second order upwind. The pressure-based solver is used with implicit formulation.

Regarding the parameters of the approach, there are six 2D cases and two 3D cases which are listed in the table 3. Therefore the final approach is composed by 8 different scenarios.

The temperature gradients are placed in the external surrounding boundaries of the BN crucible. Each node in the {0,1,0} direction corresponds to a calculated value, while the {1,0,0} direction is considered a thread of constant values (only for the 3D cases). The gradient of temperature nomenclature is called $T(y,t)$ so it depends directly on the {0,1,0} direction and time.

The top and bottom boundaries are considered a thread of cell faces with one individual value that depends only on time. $T_h$ is the hot temperature in the top of the ampoule and $T_c$ is the cold temperature considered in the bottom of it. The two gradient types are designed to consider a maximum of 100ºC of gradient and to initialize the simulation regarding the temperature of the system above the melting temperature point of the GaSb. The interesting part is to reach a pseudo-stationary state, where the growth rate is equivalent to the pulling rate of the system.

This design is done in order to be able to contrast the two gradients and in addition to stabilize the crucible T distribution on the first calculation steps, before solidification occurs.

\[
T_{lin}(y,t) = \left( T_c + y \times \frac{(T_h-T_c)}{L} \right) - pt
\]

(1)

\[
T_{lin,c}(t) = 979 - pt
\]

(2)

\[
T_{lin,h}(t) = 1079 - pt
\]

(3)

\[
p = \frac{PR}{L} \times (T_h - T_c)
\]

(4)

\[
T_{htan,wall}(y,t) = T_c + 0.5 \times (T_h - T_c) \times (1 + (htan(a \times (y - y_p)))/htan(aL \times 0.5)
\]

(5)
The equations from 1-4 and from 5-8 formalize both linear and hyperbolic tangent distribution temperatures, respectively.

\[
T_{\text{htan},c}(t) = T_c + 0.5 \times (T_h - T_c) \times (1 + (\text{htan}(a \times (-y_p))) / \text{htan}(aL \times 0.5) \quad (6)
\]

\[
T_{\text{htan},h}(t) = T_c + 0.5 \times (T_h - T_c) \times (1 + (\text{htan}(a \times (L - y_p))) / \text{htan}(aL \times 0.5) \quad (7)
\]

\[
y_p = y_{p0} + PR \times t \quad (8)
\]

The equations from 1-4 and from 5-8 formalize both linear and hyperbolic tangent distribution temperatures, respectively.

| Table 2 Values used for the T distribution formalization and other input parameters |
|-----------------------------------------------|---------------------------------|----------------|
| Parameter          | Description                                           | Value | Units |
| \( p \)          | Drop temperature rate parameter                        | 0.0014 | ºC/s  |
| \( y \)          | Each node value of the \{0,1,0\} array with L longitude | From 0 to L | m     |
| \( y_{p0} \)     | Interface initial position in \{0,1,0\} direction       | 0     | m     |
| \( a \)          | Steepness of temperature gradient in the \( \text{htan} \) eqs. | 150, 200 | -     |
| \( t \)          | Current time                                           | 0-80000 | s     |
| \( PR \)         | Pulling rate of the crucible                           | 1.4*10^6 | m/s   |
| \( L \)          | Crucible longitude in \{0,1,0\}                         | 0.1    | m     |
| \( R_i \)        | Melt/crystal radius                                     | 0.01   | m     |
| \( R_a \)        | Ampoule total radius                                    | 0.012  | m     |
| \( T_{\text{lin}} \) | Linear temperature along L distribution                | Equation 1 | ºC   |
| \( T_{\text{lin},c} \)         | Temperature on bottom of linear distribution case       | Equation 2 | ºC   |
| \( T_{\text{lin},h} \)         | Temperature on top of linear distribution case          | Equation 3 | ºC   |
| \( T_{\text{htan}} \)         | Hyperbolic tangent temperature distribution along L    | Equation 5 | ºC   |
| \( T_{\text{htan},c} \)        | Bottom temperature on the hip.tang. distribution case  | Equation 6 | ºC   |
| \( T_{\text{htan},h} \)        | Top temperature on the hip.tang. distribution case     | Equation 7 | ºC   |
| \( \Delta t \)    | Time step size (in 2D ; in 3D)                          | 0.2 ; 0.4 | s     |
| \( Num \)        | Number of time steps (in 2D ; in 3D)                    | 4*10^5 ; 2*10^5 | -   |
| \( g \)          | Gravity                                                | 10^{-4} | m/s²  |
The $y_{p0}$ is considered zero as there is no initial seed on the system. The considered boundaries for the systems are displayed in the table 4 and 5.
<table>
<thead>
<tr>
<th>Properties</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Cell num</td>
<td>1140 unrefined, 3500 refined</td>
</tr>
<tr>
<td>Mesh independency test</td>
<td>Yes</td>
</tr>
<tr>
<td>Composition</td>
<td>Quadrilateral, indexed, uniform</td>
</tr>
</tbody>
</table>
Table 5 3D mesh features

<table>
<thead>
<tr>
<th>Properties</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Cell num</td>
<td>22000</td>
</tr>
<tr>
<td>Mesh independence test</td>
<td>Yes</td>
</tr>
<tr>
<td>Composition</td>
<td>BN and outer GaSb rings: Hexahedral</td>
</tr>
<tr>
<td></td>
<td>Inner GaSb: Qua/tri hybrid</td>
</tr>
</tbody>
</table>
Numerical Method

Fluent governing equations

An enthalpy porosity method is used for the modelling of melt/solidification with the FLUENT code. The addition of a liquid fraction parameter indicates the state of the calculated cell. The liquid fraction is computed using the enthalpy balance on each iteration. The turbulence model will not be included because the velocities are very low (Re > 1).

Energy equations

The solidification/melting model by a pure has shown some peculiarities if compared with binary alloys, which consider liquid fraction by solid and liquid temperature differences [12]. The problem occurs when the solid and liquid temperature are the same, which is in the case of the pure metals. This called Stefan problem is solved by the Voller and Prakash isothermal phase change scheme [13,14]. The main features of the typical solidification/melting model are described here. Afterwards the isothermal phase change solution is explained:

The total enthalpy balance is represented as the sum of the sensible latent heats as in equation 9 $H$ is enthalpy, $h_{sens}$ is the sensible enthalpy and $\Delta H$ is the latent heat.

$$H = h_{sens} + \Delta H$$

Considering that the sensible enthalpy is (10).

$$h = h_{ref} + \int_{T_{ref}}^{T} c_p T \, dt$$

$\beta$ is considered as liquid fraction in the equation 11

$$\beta = 0 \quad \text{if } T < T_{solidus}$$

$$\beta = 1 \quad \text{if } T > T_{liquidus}$$

$$\beta = \frac{T-T_{solidus}}{T_{liquidus}-T_{solidus}} \quad \text{if } T_{liquidus} < T < T_{solidus}$$

Regarding $\beta$, the latent heat is a constrained number formalized in equation 12

$$0 < \Delta H < L_a$$

$L_a$ is the total latent heat.

The energy conservation equation in solidification/melting systems is defined as (13):
The liquid fraction cannot be calculated when a pure metal is considered, because temperature of both liquidus and solidus are the same, which produce a division by zero in equation 11.

The proposed solution by Voller and Prakash assume that the temperatures of liquid and solid are the same, so the phase change is isothermal. Equation 18 shows:

$$T_{\text{liquidus}} = T_{\text{solidus}} = T_{pc}$$

(18)

Where $T_{pc}$ is the temperature of phase change.

As for the interval $0 \leq \Delta H \leq L$ has only a single valued function. Therefore, the equation 15 becomes:

$$\Delta H^{k+1} = \Delta H^k + h^k c_p T_{pc}$$

(19)

$$\Delta H^{k+1} = \max[0, \Delta H^{k+1}]$$

$$\Delta H^{k+1} = \min[L, \Delta H^{k+1}]$$
There equation 19 is followed by the two constrains to enhance the equation 19 where $\max[a,b]$ means to look for the greater value of $a$ and $b$ and $\min[a,b]$ means the smaller of $a$ and $b$.

Eventually, if the solidus and liquidus temperature are not the same, the $f^1$ is considered as linear and the method in FLUENT documentation is used [12].

**Momentum equations**

The enthalpy-porosity method assumes the mushy region as a porous medium. This includes the source term on the Navier-Stokes equations. The added source term $S$ is displayed in the equation 20:

$$S = \frac{(1-\beta)^2}{(\beta^3+\epsilon)} \times A_{\text{mush}} \times \vec{v}$$

(20)

Where the $v$ is the velocity of the fluid, the $A_{\text{mush}}$ is a constant mushy parameter that defines the gradient of the sink of velocity of the fluid when the porosity drops ($\beta$) and $\epsilon$ is a small number (0.001) to prevent the 0 division total solidification occurs. Normally, the constant mushy parameter range is from $10^4$ to $10^7$. The greater is the value of the mushy constant the higher the velocity gradient on the interface, so this fact might provoke instabilities on the simulation is a higher value is proposed.

**Sofistications**

**Adaptative grid**

The 2D meshes have been refined two levels on the interface zone. The refinement is illustrated in the figure 3. Unfortunately, it has not been done for the 3D cases, as the mesh was configured by too many nodes and the calculation time was too high to get satisfactory results in a refined mesh.

The simulator determines which are the interface cells to be refined by using an algorithm based on determine the normalized (from values from 0 minimum to 1 maximum) gradient of the liquid fraction [12]. Once determined, those cells that present the requested gradient value are treated depending on:

- If the gradient value is lower than a threshold of 0.3, the cell is coarsened.
- If the gradient value is higher than a threshold of 0.7 the cell is refined
To transform this algorithm to a dynamic it is set that this procedure is performed every 10 time steps.

By this way, the refinement will be done in all the cells that present a gradient of the liquid fraction, so the ampoule/melt interface will also be refined increasing unnecessarily the number of nodes and therefore increasing the calculation time. The final step is to set up a maximum of refinement and coarsening level, which is defined in two steps.

**Interface Deflection**

To determine the deflection along time, a data treatment to obtain the desired results is required. The figure 4 illustrates the necessary data to determine the interface deflection and the equation 21 shows how is calculated.

\[ dh = h_s - h_c \]  

(21)
Three surfaces have been defined in order to obtain the interface deflection with FLUENT. The expression is defined in the equation 22

\[ h_c(\text{every 1000s}) = (1 - \beta(0, 0 \to L)) \times L \]  

\[ h_s(\text{every 1000s}) = (1 - \beta(\pm[R_l - 0.0001], 0 \to L) \times L \]  

The equation 23 exposes that FLUENT gathers an array of cells from 0 to L and calculates at each cell the solid fraction. Then a weighted average is performed on the array of values and finally it is multiplied by the total length \( L \). For instance:

We have an array on 100 cells in the \( \{0,1\} \) along \( L \), and FLUENT returns: 72 are in liquid phase, 27 are in solid phase and 1 has 0.5 of liquid fraction. So we obtain a final solid height of 0.275L, thus it is 0.0275m.

Whether the values of \( dh \) are positive, it means that the deflection is concave, if they are negative, means that is convex. The data is accumulated and later treated with equation 21 to display the interface deformation along time.

**Pull velocity**

Once the solid height is determined as explained in the anterior chapter, the equation 24 defines how the centre of the interface will grow with time.

\[ \frac{dh_c}{dt} \equiv PR_c \]
Parallel processing
The 3D cases have been calculated in parallel processing, meaning that the calculation time is reduced in function of the number of processors used. The machine used for the computational simulations 8-processor IC i7 CPU with 2.67GHz each processor.

Materials

Properties
The physical properties of the BN and the GaSb and other input parameters are exposed on the table 5. The boundary temperatures are defined in the equations 1-8. FLUENT is not able to compute both liquid and solid properties, so only the liquid properties of the GaSb are used as input for the simulation.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Nomenclature</th>
<th>Value</th>
<th>Units</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density</td>
<td>$\rho_{\text{GaSb}}$</td>
<td>6060</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td></td>
<td>$\rho_{\text{BN}}$</td>
<td>1900</td>
<td>kg/m$^3$</td>
</tr>
<tr>
<td>Thermal Conductivity</td>
<td>$k_{\text{GaSb}}$</td>
<td>10.24</td>
<td>W/(m·K)</td>
</tr>
<tr>
<td></td>
<td>$k_{\text{BN}}$</td>
<td>20.6</td>
<td>W/(m·K)</td>
</tr>
<tr>
<td>Specific Heat</td>
<td>$c_{p\text{GaSb}}$</td>
<td>330</td>
<td>J/K·kg</td>
</tr>
<tr>
<td></td>
<td>$c_{p\text{BN}}$</td>
<td>1850</td>
<td>J/K·kg</td>
</tr>
<tr>
<td>Latent Heat</td>
<td>$L_a$</td>
<td>313531</td>
<td>J/kg</td>
</tr>
<tr>
<td>Thermal diffusivity</td>
<td>$\alpha$</td>
<td>5.1*10$^{-6}$</td>
<td>m$^2$/s</td>
</tr>
<tr>
<td>Melting point</td>
<td>$T_m$</td>
<td>979</td>
<td>K</td>
</tr>
<tr>
<td>Viscosity of melt</td>
<td>$\mu$</td>
<td>2.242*10$^{-3}$</td>
<td>Kg/m·s</td>
</tr>
<tr>
<td>Thermal expansion coefficient</td>
<td>$\beta_T$</td>
<td>10$^{-4}$</td>
<td>K$^{-1}$</td>
</tr>
<tr>
<td>Total effective gradient of temperature</td>
<td>$\Delta T_s$</td>
<td>100</td>
<td>K</td>
</tr>
<tr>
<td>Gravity</td>
<td>$g$</td>
<td>10$^{-4}$</td>
<td>m/s$^2$</td>
</tr>
</tbody>
</table>
Nondimensional numbers

The used nondimensional Grashof and Prandtl numbers are defined as in equation 25 and 26

$$Gr = \frac{\rho^2 g \Delta T \beta R_i^3}{\mu^2}$$  \hspace{1cm} (25)

$$Pr = \frac{\mu c_p}{k}$$  \hspace{1cm} (26)

Where \(\nu\) is the kinematic viscosity in m\(^2\)/s. The Gr and Pr values on the steady state are very low in all scenarios (Gr <100, Pr = 0.07) which means that the fluid is laminar at the ampoule/interface layers and also that GaSb is a strong conductor.

3. Results and Discussion

The results regarding the linear and \(htan\) distributions are displayed in this chapter. A contrast between different gravity vector orientations will be done to assess if there exist changes in the interface during the crystal growth.

The results in 3D are not fully calculated yet, so only the finished cases are displayed, which are the two linear and \(htan\) in the vertical gravity vector. The velocity of the fluid, the stream function and the interface are determined, as well as the direction of the fluid currents.

The contrast will be summarized by displaying the interface deflection in the three different gravity directions and with the two different temperature distributions in two separated graphics. It is expected that velocity on the crystal/melt interface will be greater in the \(htan\) temperature distribution because the whole concentration gradient (100K) is localized mainly in the interface.

Prior to this contrast, the T and grid evolutions on time are displayed, as they do not present significant differences from one case to another when the orientation of the gravity vector is changed.

Temperature distribution and adaptable grid

\(T\) distribution

After obtaining all the results, it has been observed that T distributions are the similar regardless the orientation of the gravity vector. The T distribution is shown by isotherm maps...
of 10 degrees Kelvin each isotherm. The figures 5 and 6 represent the T distributions in the linear and \( \text{htan} \), respectively. The table 7 gives the maximum and minimum temperature values for the linear case. The interface is pointed in each figure. A table of maximum and minimum is not included for the hyperbolic tangent distribution as the maximum and minimum relies always from 929 to 1029K.

![Figure 5](image)

**Figure 5** General linear T distribution in different times: a) 6000 b) 18000 c) 30000 d) 42000

<table>
<thead>
<tr>
<th>in K</th>
<th>6000s</th>
<th>18000s</th>
<th>30000s</th>
<th>42000s</th>
</tr>
</thead>
<tbody>
<tr>
<td>Max</td>
<td>1070</td>
<td>1054</td>
<td>1039</td>
<td>923</td>
</tr>
<tr>
<td>Min</td>
<td>970</td>
<td>954</td>
<td>939</td>
<td>1023</td>
</tr>
</tbody>
</table>

Table 7 Maximum and minimum T values for lineal distribution cases in different times
Figure 6 General $htan$ T distribution in different times: a) 6000 b) 18000 c) 30000 d) 42000

Figure 5a has a gradient of temperature between isotherms of 7 degrees Kelvin because the gradient is progressively performed in the $htan$ cases to prevent instabilities on the initial steps. The figures 6b to 6d have the same thermal gradient from 929 to 1029K. The interface is pointed with arrows, corresponding to a T of 979K.

Adaptable grid

Figure 7 illustrates how the mesh is simultaneously refined and coarsened to obtain the maximum node density in the interface to help to improve the quality of the simulations.
Figure 7 shows how the absolute quantity of nodes is decreases with time because the number of cells that need to be refined is lower. The melt/ampoule interface, which is liquid/solid interface, decreases whereas the crystal/ampoule interface, which is solid/solid, increases the cell number. We need to remember that the refinement algorithm takes in account the melt/ampoule interface to refine even if it is necessary for this simulation. So this means that the calculations will go faster when the time passes and the crystal/melt interface grows.
2D scenarios

*Linear T distribution cases: vertical, horizontal and diagonal*

*Interface and flow evolution*

![Interface and flow evolution](image)

**Figure 8** Interface: linear T, vertical gravity: a) 6000s b) 18000s c) 30000s d) 42000s

![Stream function](image)

**Figure 9** Stream function: linear T, vertical gravity: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 10 Velocity vectors: linear T, vertical gravity: a) 6000s b) 18000s c) 30000s d) 42000s

Figure 11 Interface: linear T, horizontal: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 12 Stream function: linear T, horizontal: a) 6000s b) 18000s c) 30000s d) 42000s

Figure 13 Velocity vectors: linear T, horizontal: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 14 Interface: linear T, diagonal: a) 6000s b) 18000s c) 30000s d) 42000s

Figure 15 Stream function: linear T, diagonal: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 16 Velocity vectors: linear T, diagonal: a) 6000s b) 18000s c) 30000s d) 42000s

\textit{Htan T distribution cases: vertical, horizontal and diagonal}

Figure 17 Interface: \textit{htan T}, vertical: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 18 Stream function: $htan T$, vertical gravity: a) 6000s b) 18000s c) 30000s d) 42000s

Figure 19 Velocity vectors: $htan T$, vertical gravity: a) 6000s b) 18000s c) 30000s d) 42000s
**Results and Discussion**

**Figure 20** Interface: linear $T$, horizontal: a) 6000s b) 18000s c) 30000s d) 42000s

**Figure 21** Stream function: $htan\ T$, horizontal: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 22 Velocity vectors: $htan T$, horizontal: a) 6000s b) 18000s c) 30000s d) 42000s

Figure 23 Interface: $htan T$, diagonal: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 24 Stream function: $htan \ T$, diagonal: a) 6000s b) 18000s c) 30000s d) 42000s

Figure 25 Velocity vectors: $htan \ T$, diagonal: a) 6000s b) 18000s c) 30000s d) 42000s
In table 8 are the given values of the stream function of the 2D cases. There exists a relation between the stream function and the interface deflection, as the higher are the stream function, the higher is the Gr number and high Gr values are not favourable to keep a flat interface.

<table>
<thead>
<tr>
<th>Gravity orientation</th>
<th>6000s</th>
<th>18000s</th>
<th>30000s</th>
<th>42000s</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linear Distribution</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vertical</td>
<td>3.26*10^-7</td>
<td>4.63*10^-7</td>
<td>4.60*10^-7</td>
<td>4.60*10^-7</td>
</tr>
<tr>
<td>Horizontal</td>
<td>6.92*10^-5</td>
<td>6.92*10^-5</td>
<td>6.92*10^-5</td>
<td>6.72*10^-5</td>
</tr>
<tr>
<td>Diagonal</td>
<td>4.09*10^-5</td>
<td>4.89*10^-5</td>
<td>4.90*10^-5</td>
<td>4.80*10^-5</td>
</tr>
<tr>
<td>Hyperbolic tangent</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Vertical</td>
<td>1.54*10^-5</td>
<td>1.35*10^-5</td>
<td>1.36*10^-5</td>
<td>1.36*10^-5</td>
</tr>
</tbody>
</table>

The given figures 8-25 show up how the flow and interface evolve on time. The VB systems make move the flow by creating two symmetric swirl currents on touch with the interface figure 8-9 and 18-19. The VB with linear T configuration gives the lowest stream function values among all the simulated configurations.

By comparing the 8-9 and 18-19 there is an inversion of the current direction. Whereas in the figures 8-9 swirl on the left goes in counter-clockwise direction, the other swirl in figure 18-19 goes clockwise, which means that the flow goes downwards in the centre. This phenomenon is directly related to the hyperbolic T distribution which has a steep temperature gradient only on the surface. There exist many references that experiment this swirl inversion in different conditions [5,16,17], most of them in accelerated crucible rotation techniques. None of the references explain explicitly how this phenomenon occurs. In this framework the inversion of the currents is produced at 5-6 seconds spontaneously.

Another observation is that there is an important variation between the linear and hyperbolic T distribution when assessing the stream function shape of the figures 12-13, 15-16, 21-22, 24-25 which correspond on all the horizontal and diagonal cases. While in the stream function on the linear distribution in diagonal and horizontal gravity orientations assembles to the ampoule hole, in the hyperbolic T case the stream function is localized in the melt/crystal interface. This phenomenon is justified because the gradient of the linear T distribution is
CRYSTAL GROWTH INTERFACE AND MICROGRAVITY

along the whole lateral walls, and the \( htan \) \( T \) gradient only occurs in a localized part that conforms the 20-30% of the lateral walls.

**Interface deflection: linear and \( htan \) contrast**

The figure 26 illustrates the evolution of the interface deflection with time.

![dh vs t: Linear T distribution scenario: V, H and D gravity vector orientations](image)

**Figure 26** \( dh \) vs \( t \): Linear \( T \) distribution scenario: V, H and D gravity vector orientations

The interface deflection is the same for the three cases except the first 8000s, and stabilizes at approximately \( 7.4 \times 10^{-4} \) m after the 15000 seconds. This common deflection value for the three gravity orientations means that interface deflection does not depend on the orientation of the gravity vector.

The visible fluctuations that show the figure 26 are typical from a not enough refined mesh and therefore the interface deflection does not fluctuate in reality.

The interface deflection for the \( htan \) \( T \) distribution cases is illustrated in the figure 27. The first transient 15000 seconds the deflections has negative values, thus, the interface is convex.

According with C.W. Lan work [18], this phenomenon is enhanced with localized high gradients. The approximate value of the stabilized deflection in the \( htan \) scenarios is around \( 3 \times 10^{-4} \) m, which is less than the half than on the linear scenario. So, it is evident that the countered flow of the swirls and the initial convexity of the interface take part to improve crystal segregation for this system. Actually, it is proven that the direction swirls comes back upwards on the centre when the \( dh_c \) reaches a critical value.
Results and Discussion

Crystal Growth Interface And Microgravity

**Figure 27** $dh$ vs $t$: $H_{tan}$ $T$ distribution scenario: V, H and D gravity vector orientations

### 3D cases

The two vertical cases are exposed in this chapter. The stream function cannot be displayed as it is only available for 2D systems. Also need to be regarded that the mesh here cannot be refined.

**Interface and flow evolution**

![Diagram showing interface and flow evolution](attachment:image.png)

**Figure 28** Interface: 3D, linear $T$, vertical, xy plane: a) 6000s b) 18000s c) 30000s d) 42000s
Figure 29: Velocity vectors: 3d, linear T, vertical, xy plane: a) 6000s b) 18000s c) 30000s d) 42000s

Figure 30: Velocity vectors: zoom of figure 29b
Results and Discussion

Crystal Growth Interface and Microgravity

**Figure 31** Interface: 3d, $htan\ T$, vertical, xy plane: a) 6000s b) 18000s c) 30000s d) 42000s

**Figure 32** Velocity vectors: 3d, $htan\ T$, vertical, xy plane: a) 6000s b) 18000s c) 30000s d) 42000s
The 3D vertical cases lead to the same observations as the 2D approach. There is an inversion of the swirl flows and the melt flows downwards (figure 33) in the centre of the crucible when the $htan$ T distribution is applied. This fact enhances the theory that the system regarding the specified boundaries creates countered flows.

The interface evolution illustrated in the figures 28 and 31 shows some abrupt deformations of the interface. Again, these deformations exist due to the low density of the mesh. The deformation magnitude is equal to the size of the cell length.

**Interface deflection: linear vs $htan$ T distribution**

The figure 34 illustrates the interface deflection of the 3D linear and $htan$ T distribution. According to the interface deformations presented in the figures 28 and 31 and contrasting this graph with figure 27, the fluctuations are notably higher. Nevertheless, these fluctuations still are produced around similar values of the 2D vertical cases, which are around $9*10^{-4}$m for the linear T distribution case and $5*10^{-4}$m for the $htan$ T distribution case.
Figure 34 $dh$, along time: 3D, linear $T$ distribution, vertical gravity

Figure 35 $dh$, along time: 3D, $\tan T$ distribution, vertical gravity
4. Conclusions and outlook

1. The simulation of a Bridgman directional crystallization is satisfactorily done in 2D and 3D approaches.
2. The interface refinement procedure has been a useful tool to accurately detect the shape of the interface in all the calculated 2D cases.
3. The results obtained from this work reinforce that the hypothesis of flat interface is valid in all the analyzed cases.
4. In the present simulations, the interface velocity tends, in the pseudo-stationary state to the given pulling velocity. This is because the crystal solidification in the centre has time to occur at the same rate as the pulling one.
5. The low gravity conditions make the g-orientation to be irrelevant when the interface wants to be controlled. However, the temperature distribution still takes an important role on developing the flow of the melt.
6. Hyperbolic temperature boundary condition seems to decrease the interface deflection compared to the lineal temperature boundary distribution.

Nonetheless, the single geometry and gravity values used in the present study are only a first approximation to accurately predict real microgravity conditions. The next upcoming issues that are likely to be regarded in the following recommendations for this project are:

- **Finish the 3D approach**: to be able to contrast the diagonal and horizontal g-oriented 2D and 3D cases.
- **Variations of Gr and Pr numbers**: physical property changes on both ampoule and semiconductor, geometry, characteristic length of the interface and gravity magnitude.
- **Implementation of concentration model**: develop a model containing within the calculations a dopant and to assess the concentration distribution along the crystal.
- **Improvement of energy model**: by adding mainly the radiation model and the wall resistance model.
- **Add a seed to the system**: the system would reduce the transition period and there would increase the effective time in stationary state, which is the desired state.
- Represent more realistic effect such as residual accelerations (g-jitter): to include a realistic orbit effects to the model and assess how these forces affect on the interface, concentration distribution and heat and mass flow.
Acknowledgements

The project would not have been done without the contribution of Josep Xavier Ruiz who has been my mentor in the topic of crystal growth and provided me the information and the required hardware to perform the simulations. I would like to thank Jordi Pallarès for helping me in some critical parts of the project, especially, on the computational problematic, which has demonstrated that he is an expert on the topic.
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[12] ANSYS Fluent 13.0 theory guide, chapter 18, 2010


