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COMPUTATIONAL MODELING OF HEAT AND MASS TRANSPORT AND CONTROL OF SOLID/LIQUID TRANSITION SYSTEMS

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Abstract. *This paper deals with a review of numerical investigations of solid/liquid phase change configurations obtained by using an enthalpy-porosity formulation coupled to a finite volumes approximation. Attention is focused on the control of convective motion in the liquid phase using operating parameters as growth rate and thermal gradient of the furnace. Situations involving thermal convection holding in the earth and in the microgravity environment are discussed. Control using constant magnetic field is illustrated.*

1 INTRODUCTION

In crystal growth devices, in order to control the quality of the obtained product, a thorough understanding of the heat transfer characteristics is required to optimize the design and the operating conditions of the process. Computational modeling and simulation give a good way for interaction with experiments. Developed codes shall be able of accurately calculating flow, temperature and concentration field and to take into account multiple involved scales.

Regarding the solid/liquid phase change with free or moving boundary conditions increases the complexity of classical fluid mechanic configurations and requires specific Computational Fluid Dynamics/Heat Transfer models. The aim is to contribute to a better understanding of complex structures in fluid flow and to propose efficient control ways to increase the quality of grown products [1, 2].

The convective motions affect transfer in the vicinity of the solidification interface and are subject to many studies. This instability is coupled to solidification front motion. The interfacial area is often very complex to describe. Indeed, front curvature and solid phase structure depend on several factors and it will be interesting to eliminate some of them due to the gravity for a best understanding of solidification process. Experiments in microgravity conditions are achieved, in orbiting space platforms, parabolic flight, sounding rockets and satellites [4, 5]. Control process by vibration or magnetic field are sometimes required to stabilise solidification fronts [6, 13].

The present contribution summarizes numerical results of convection arising in the solidification and discuss the control of such configurations by dumping the convection. Illustrations focuses on Bridgman and Floating Zone processes commonly employed to grow single crystals especially for electronic industry. Examples of magnetic field use, microgravity environment effect and operating furnace parameters influence are presented.

2 SOURCES OF CONVECTION

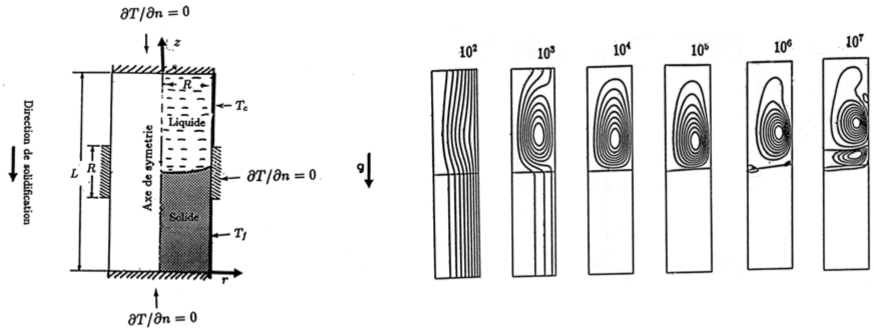
There can be several driving forces for convection in phase change systems due to the gravity, the thermal and density gradients caused by compositional homogeneities. Depending on the mutual orientation of gravity and such gradients, buoyancy driven convection can be set up and alter the scalar fields and also the solid/liquid interface shape. If there is a free surface separating liquid and gas domains, a surface tension usually will exists leading to the Marangoni convection. Theses different factors can give very complex patterns when interacting. Many other modes of convection can exist and interact, such as forced one due to crystal rotation, time varying electromagnetic field [1-3, 7, 8, 11].

3 GROWTH ENVIRONMENT AND CONTROL

During crystal growth from the melt in a crucible, thermal transfer in the furnace/crucible/sample systems lead to curvature of the isotherms and specially of the interface and is detrimental of crystal quality. Several strategies are proposed in order to limit the curvature of the solid/liquid interface and are generally based on operating parameters as growth rate and global heat transfer or on environing fields as the gravity or magnetic fields.

3.1 Microgravity environment

The electronic properties of the crystal grown by directional solidification are critically dependent on the distribution of the trace elements within the crystal. Convection can alter this distribution and may degrade the quality. The space laboratory and recent shuttle experiments indicate that it is possible to grow crystals without buoyancy driven convection. The other interest of microgravity conditions is the possible separation of each process such as a radiation from the effect of buoyancy driven. In the earth all processes happen at the same time [4, 5, 10]. Figure 1 concerns the effect of Ra on fluid flow showing transition from diffusion regime to mulicellular one (fig. 1(a)). The given illustration concerns a Gallium doped Germanium growth (Ga-Ge) corresponding to a Prandtl number $Pr=\nu/\alpha=10^{-2}$ (ν is the kinematics viscosity). The configuration is vertical Bridgman one. The translation of the ampoule in the furnace is considered by using a Peclet number $Pe=Vt/V_{ref}=10^{-2}$ corresponding to a constant growth rate of $V_t=4$ µm/s (V_{ref} is the reference velocity). The ampoule is cylindrical with aspect ratio A=2 but calculations are achieved in axisymmetrical configuration. The effect of gravity is considered via the Rayleigh number based on constant ΔT, $Ra=g\beta\Delta TL^3/\nu\alpha$ (α is the thermal diffusivity of the liquid phase). The Latent heat of fusion (L) is taken into account via Stefan number $Ste=c\Delta T/L=1$ (c is the heat capacity of the liquid phase).



(a) (b)

Figure 1. Melt under directional solidification using vertical Bridgman method (a) for $10^2 \leq Ra \leq 10^7$ (i.e gravity effect) showing evolution from mono-cellular to multicellular flow (b). Simulations correspond to an enthalpy- porosity formulation/FiniteVolumes method [8].

3.2 Magnetic field

In real microgravity experiments, gravity magnitude could not completely suppressed and generally a small but not negligible thermo-convective flow subsists. The near environment could also disturb the microgravity conditions and induces unsteady flows (G-jitter phenomena) [9]. An alternative solution to dump natural convective motions during a crystal growth process is the use of a magnetic field parallel to the axis of the ampoule and opposite to the gravity vector. The action of Lorenz force upon the fluid particle induces a significant dumping of flow in the melting zone. These technique represents a promising way to perform high quality single crystals both on earth and on space. An example of dumping convection using magnetic field is presented in the Figure 2. The same configuration described in the previous part is considered and the constant vertical magnetic field on the electrically conducting fluid is considered via the Loretz body Force involving Hartman dimensionless number $Ha=(\sigma BL^2/\mu)^{1/2}$($\sigma$ is the electrical conductivity, μ is the dynamic viscosity and B the magnetic field magnitude).

The results obtained by increasing magnetic field effect (Ha) show that the flow pattern is crushed along the ampoule wall and damped by viscous and Lorentz force effect. The convection in the multicellular flow corresponding to $Ra=10^7$ is progressively dumped by increasing Ha showing the opposite effect of the magnetic field.

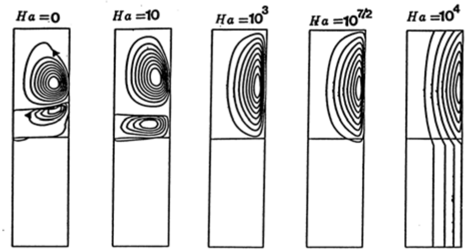
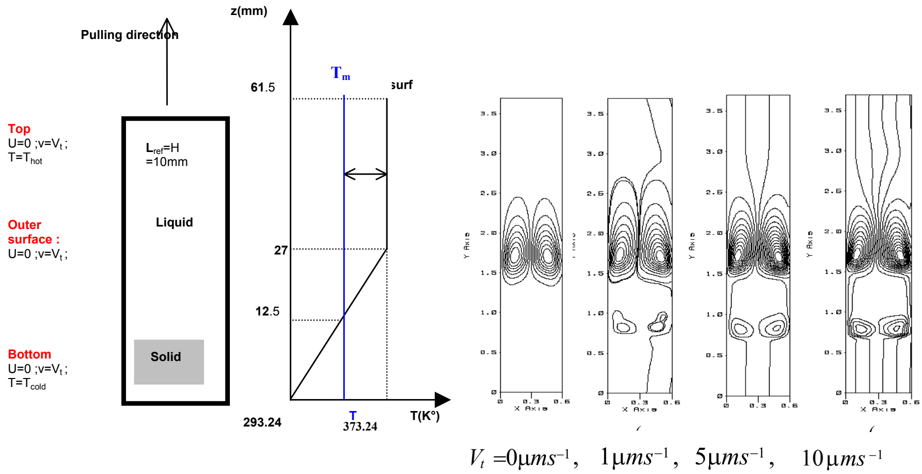


Fig : Effect of constant magnetic field ($0 \leq Ha \leq 10^4$) opposite to gravity on multi-cellular flow corresponding to $Ra=10^7$ for vertical Bridgman configuration [8].

3.3 Growth velocity



(a) (b)

Figure 3. (a) Directional solidification of SCN-Acetone. (b) streamline plots of solutal convection corresponding to $0 \leq Pe \leq 10$ from left to right [12].

The simulation is carried out for the configuration sketched in the figure 3 (a) and concerns Bridgman vertical growth of SCN-Acetone (Pr=22.8, Ste=1.71) under full gravity conditions $Ra=1.12\ 10^6$ and for four pulling rates. For no growth rate quasi uniform thermal field is observed and the melt is occupied by two counter rotating cells in good agreement with classical numerical results for such configurations [8, 12]. This flow is driven by the radial temperature gradients established by transition in the thermal boundary between the gradient zone and the hot zone.

Increasing the pulling velocity, the melt-crystal interface becomes concave. Two counter–rotating secondary convective cells next to the interface are in opposite direction to the main cells and occupy a more important space domain. It is noted that the maximum stream function increases and convection has a flattening effect on the isotherms in fluid phases as V_t increases ($\Psi_{max}=0.267 mm^2 s^{-1}$ for $V_t=1\mu ms^{-1}$, $\Psi_{max}=0.293 mm^2 s^{-1}$ for $V_t=5\mu ms^{-1}$ and $\Psi_{max}=0.346 mm^2 s^{-1}$ for $V_t=10\mu ms^{-1}$).

3.4 Furnace boundary conditions

At the entire system level, the thermal environment affect the global growth via heat transfer through the ampoule walls. An optimization of growth morphology (interface shape, constitution, …) can depends on heating conditions.

When a crystal growth configuration is submitted to an external heat flux, the hydrodynamic of the problem depends strongly on the applied flux distribution. An example of Floating Zone simulation shows how this external parameter is important for the quality of the obtained crystal by FZ process. Results show the effects of constant and polynomial distribution on velocity field and interface with fixing the Marangoni number value ($Ma=-\frac{\partial\gamma}{\partial T}\Delta T\, R/\mu\alpha$, γ is the surface tension and R the cylindrical ampoule radius) [14].

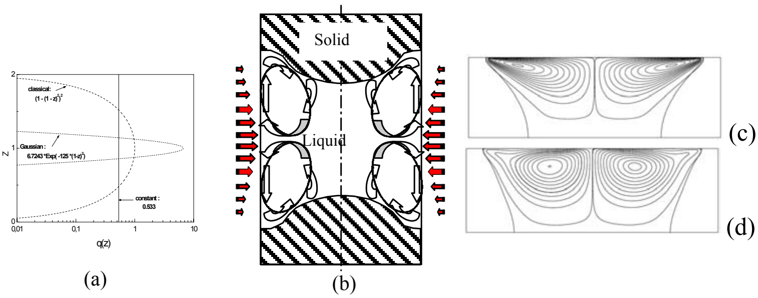


Figure : Floazing Zone configuration (b) controlled using thermal boundary conditions (a). The flux concentration effect on the temperature and the flow field for constant (c) and Gaussian (d) applied flux (Ma = 5.10^3, Pr = 1.0). For constant flux ($|\psi_{max}|$=2.18 mm^2/s) and for Gaussian flux ($|\psi_{max}|$=12.52 mm^2/s) [14].

4 DISCUSSION

Computational results of this paper are essentially obtained using enthalpy-porosity formulation and finite volume numerical approximation. Various illustrative cases show that the approach could treat such melting/solidification problems without continual remeshing of the domain that follow the movement of the fusion solidification front. The main illustration concerns the possible control of grown products. Microgravity or magnetic field environment allows a global control to dump convection in the melt. The translation rate (pulling velocity) and heating conditions allows a direct control on operating conditions.

The work follows on providing schemes for optimizing process with combining external and internal control parameters and enlarged to rotating flows and Marangoni convection of growth.

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