

3D computer simulation of convective instability in the multi-component solution

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Abstract: -The paper presents the results of numerical study of natural convection in multicomponent solution. Three-dimensional calculations have been done to simulate the onset of convective motion and correspondent flow patterns. The process is considered for Rayleigh number in the range $1 \cdot 10^3 \div 4 \cdot 10^4$, where subcritical convective motion with hexagonal flow pattern is identified. Also transition from regular cellular convection to to skewed - varicose and knot instability is registered. The results are in good agreement with the linear and finite amplitude theory of hydrodynamics stability.

Keywords: convective instability, subcritical convection, computer simulation.

1 Introduction

Numerical study of mass-transfer in multicomponent systems with phase transition is one of the most challenging problems for computational fluid dynamics. The investigations in this field are encouraged by the requirements of the modern crystal growth technology.

The paper deals with computer simulation of natural convection in solidification of ternary nondilute solution. The problem arises in computer simulation of liquid phase epitaxy (LPE) – a crystal growth technique that is widely used for preparation of multicomponent semiconductor materials with desired structural composition [1].

The basic principle underlying the growth of epitaxial layers by LPE is similar to that of growth of salt crystals from a saturated saline solution [2]. In a typical setup a saturated solution of the components A and B in molten C is brought into contact with a substrate $A_xB_{1-x}C$. For definiteness, the substrate is supposed to be placed horizontally under the solution.

To initiate and sustain the growth the system temperature is decreased. Due to the cooling, initially saturated solution becomes supersatu-

rated; components A, B precipitate out of the solution onto the substrate. The mixture is then relaxed to equilibrium, forming the solid and the saturated solution. At the solid – liquid interface, compositions of the produced two phases satisfy the phase diagram, while the bulk of the liquid retains supersaturated. The reduction in the concentration of growth units in the vicinity of the growing layer gives rise to nonlinear concentration profile and leads to density gradient normal to the substrate. It can be stable, if the solvent has a greater density than the solutes, and unstable, if the densities are differently disposed. Here we consider the last case and suppose both components A and B contribute to instability. It means that solutal unstable density gradient provides a driving force for convective motion. This is the principle convective mode in LPE. A second mode may occur due to the gradual temperature changes.

The non-dimensional parameters, characterizing the relative strength of buoyancy forces in natural convection, are solutal (Ra_{D_i}) and thermal (Ra_T) Rayleigh numbers. They are induced by concentration and temperature gradients respectively: $Ra_{D_i} = \frac{g\beta_i H^3 \Delta C_i}{D_i \nu}$, $Ra_T =$

$\frac{g\beta_T H^3 \Delta T}{\kappa\nu}$, where $i = A, B$, g is the gravitation constant, β_i is solutal expansion coefficient, H is the characteristic dimension, ν is the kinematic viscosity, D_i is the diffusion coefficient, ΔC_i is the characteristic concentration difference, κ is the thermal diffusivity, β_T is thermal expansion coefficient, and ΔT is the characteristic temperature difference.

For reasonable operating conditions, solutal Rayleigh number varies in the range $Ra_D \sim 10^3 \div 10^5$. The thermal Rayleigh number, due to slow cooling rate and good heat conductivity of the liquid phase, is far lower than solutal one: $\frac{Ra_{D_i}}{Ra_T} \sim 10^2$ [2]. It means that thermal convection is negligible. The temperature distribution is approximately uniform throughout the system, but its value changes with time. These temperature changes do not affect the fluid flow and solute transport significantly. They displace the equilibria and alter the phase relations at the crystal-liquid interface [3, 4].

It is well recognized now that transport phenomena in the solution have a profound influence on the property of the growing epitaxial layer [4, 5]. Thus the comprehensive study of solutal convection is crucial for the optimization and refinement of the technique.

2 Mathematical model

Computer simulation for LPE growth of $A_x B_{1-x} C$ structures is based on the following assumptions. The growth takes place under quasiequilibrium conditions. In particular, it means that, at the solid-liquid interface, the composition of the two phases are related by phase diagram of the system. Temperature throughout the system is supposed to be uniform in space and changes in time according to the prescribed rule: $T(t) = T_0 - \alpha(t)t$, where T_0 is the initial temperature, $\alpha(t)$ is the cooling rate and t is time.

The surface kinetics effects are ignored, the growth rate is not limited by the interface phenomena and controlled by bulk transport. Mass transfer in the solution is determined by diffusion

and natural convection. Diffusion in the solid phase is neglected. Furthermore, as the thickness of epitaxial layer is usually within 1 – 2% of the solution depth, the change of the liquid volume caused by the film growth is neglected. The solution is supposed to be incompressible and Boussinesq approximation is adopted.

Under the above assumptions, the process is described by 3D time-dependent fluid flow and mass transport equations.

In Cartesian coordinates (x, y, z) , the dimensionless governing equations take the form

$$\begin{aligned} \partial_t V + (V \cdot \nabla)V &= -\nabla p + \Delta V + \sum_{i=1}^2 Gr_i C_i e_z \\ \nabla V &= 0 \\ \partial_t C_i + (V \cdot \nabla)C_i &= \frac{1}{Sc_i} \Delta C_i \end{aligned} \quad (1)$$

where $\partial_\xi \equiv \frac{\partial}{\partial \xi}$, $\nabla = (\partial_x, \partial_y, \partial_z)$, $\Delta = \nabla^2 = \partial_{xx}^2 + \partial_{yy}^2 + \partial_{zz}^2$, $(x, y, z) \in \mathcal{D}$, $\mathcal{D} = [0, L] \times [0, L] \times [0, H]$, $L/H \approx 10$, $V = (V_x, V_y, V_z)$ is the velocity vector, p is pressure, C_i , $i = A, B$ is the concentration of the correspondent component dissolved in the liquid phase, $Gr_{D_i} = \frac{g\beta_i H^3 \Delta C_i}{\nu^2}$ is the solutal Grashoff number, $Sc_i = \nu/D_i$ is the Schmidt number, $e_z = (0, 0, -1)$.

The non-dimensional variables are introduced by scaling the length with the depth of the liquid phase (H), time is scaled with H^2/ν and concentration is scaled with the initial concentration of one of the components in the solution.

The phase transition is going on the substrate that is placed under the solution at $z = 0$. The interface conditions on concentration fields consists of the mass balance between the transported and incorporated solute species:

$$V_{gr} = \frac{\partial_n C_A}{Sc_A(C_A^s - C_A)} = \frac{\partial_n C_B}{Sc_B(C_B^s - C_B)} \quad (2)$$

and phase diagram representing the equilibrium between the solution and growing layer:

$$F_{liq}(C_i, T) = 0, \quad F_{sol}(C_i^s, C_i, T) = 0 \quad (3),$$

where $T(t)$ - temperature of the system, C_i^s - concentration of the correspondent solute specie in

the solid phase. On the remainder of the boundary: $\partial_n C_i = 0$. Boundary condition on velocity field is $V = 0$.

At $t = 0$ the liquid is in the static state. Initial condition for concentration is determined by phase diagram at temperature T_0 and the substrate composition with which the liquid is in equilibrium.

3 On the finite amplitude instability

The onset of convection in LPE system has a known so far analogy. Neglecting the existence of the component with smaller Grashoff number, let it be component B, and supposing linearity of phase diagram, we actually obtain the problem concerning convective instability in a horizontal fluid layer with nonlinear undisturbed profile of active scalar quantity (concentration).

The main results concerning the problem goes back to [6, 7, 8]. R. Krishnamurti has investigated the onset of convection in the infinite horizontal layer with the mean temperature, as an active quantity, changing steadily at a rate η . This represent the case in which the conduction temperature profile is parabolic, its shape is independent of time, while all points in the liquid change in temperature at the same rate η . The sign and the absolute magnitude of η define the direction of the profile curvature and the curvature itself respectively.

In [7, 8] it has been shown that the nonlinear conductive temperature profile, forming in the fluid when the mean temperature changes at a rate η , gives rise to stable hexagonal flows for a range of Rayleigh numbers near the critical. The direction of flow at the center of the hexagon is downward if η is positive and upward if η is negative. The static state is unstable to finite amplitude disturbances below the critical point Ra_{cr} predicted by linear theory. In particular, for non-dimensional value $|\eta| \sim 5 \div 10$, a change up to 40% from the critical number has been established.

In epitaxial growth, the role of active scalar quantity instead of temperature plays concentra-

tion. Its evolution at the interface is governed by the parameter $|\tilde{\eta}| = |k\alpha|$, where k is the slope of phase diagram, α is a cooling rate; $\tilde{\eta}$ can be treated as the temperature changing rate η in [7]. For a practically used operating conditions [5, 9] $|\tilde{\eta}| \sim 3 \div 8$. (The scaling coincides with [7].)

Though $\tilde{\eta}$ varies with time and the process is time-dependent, it is interesting to obtain Rayleigh number for the onset of convective motion, i.e. the Rayleigh number beyond which the steady state would become unstable. The result also would have a significant practical value because diffusion limited mass transfer in liquid phase epitaxy is favorable for the material quality [5, 9]. For this purpose within the scope of the model described above, direct numerical simulation of the process has been carried out.

4 Numerical procedure

The governing equations (1) are approximated at staggered grid using control-volume approach. Discretization is conservative of kinetic energy and concentration of the dissolved species. The scheme is implicit, has second order in space and first in time. Navier - Stokes equations and equations of mass transfer are solved successively at each time level.

To determine velocity and pressure fields we use a projection method. In the calculation of the predicted velocity field, the pressure gradient at the previous time step is taken into account. The projection step is performed by solving a Poisson problem for a pressure-correction.

The calculated velocity field is inserted into the mass transfer equations. Being independent inside the region, they are coupled at the solid-liquid interface by mass balance conditions (2) and phase diagram (3). The attempts to decouple them in any sequential procedure or by iteration produce highly unreliable procedure with poor if any convergence [10]. To overcome the difficulties, we use the algorithm based on coupled solution of mass transfer equations with respect to the concentration of both species and Newton method to deal with nonlinearity in boundary conditions. The linear systems involved at each Newton iteration are solved by

conjugate gradient method. The approach has been used successfully in 2D case [5, 9] and was extended to 3D.

5 Numerical results

Full scale computer simulation for LPE growth under reasonable operating conditions has been done for Rayleigh number $Ra = \max_i Gr_i Sc_i$ varying in the range $1.1 \cdot 10^3 < Ra < 3.5 \cdot 10^4$, $Sc_i = 50$.

Fig.1 schematically illustrates time evolution of undisturbed concentration field from the homogeneous initial data to a parabolic profile. The difference between the concentration of the solution at the top and at the bottom increases in time as well as the curvature of the concentration profile. Both factors are favorable for the beginning of convection.

Calculated time history of Rayleigh number for $H = 1.1$ is shown in fig.2. The time averaged value of $\tilde{\eta}$ is -5.

The simulation presents a transition from static state to non-regular flow pattern, then to the rolls and at last to the hexagonal planform (Fig.3). The motion is upward in the center of hexagon and down along the sides. That is the so called cell of "l"-type [11]. The run duration is about 25 vertical diffusion times $t_{D_i} = H^2/D_i$.

The onset of convective motion is observed at $Ra = 1100$, that is approximately 40% less than well known value 1708 predicted by linear stability theory for horizontal layer with rigid boundaries and linear profile of active scalar quantity.

The result exactly agrees with R.Krishnamurti finite amplitude stability analysis [7, 8]. In subcritical region, hexagons are the only stable finite amplitude convection. When the Rayleigh number is slowly increased the convection starts growing near the critical Rayleigh number in the form of rolls. For nonlinear undisturbed profile, this flow pattern is unstable and convective motion settles at the finite amplitude value of the stable hexagon solution. For $\eta < 0$ it should be hexagonal cells of "l"-type.

Computational results for higher Rayleigh numbers are presented in fig. 4-6. In the range

$1.1 \cdot 10^3 < Ra < 1.4 \cdot 10^4$, a regular cellular convection pattern is obtained.(Fig.4) For Rayleigh number between $1.4 \cdot 10^4$ and $4 \cdot 10^4$ the transition to skewed-varicose and knot instability is registered [11] (Fig.5,6). The mean convective structure size increases with Ra while $Ra < 3 \cdot 10^4$. Above $Ra = 3 \cdot 10^4$ chaotic cellular small-scale convection is detected (Fig.6). The mean cell size at $Ra = 3.5 \cdot 10^4$ is several times less than the case of $Ra = 1.8 \cdot 10^4$.

The same transition in fluid flow structure under the increased Rayleigh numbers was observed in experimental study for onset and development of convective motion in horizontal layer with internal heat generation [12]. That is another classical for the stability analysis problem similar to the considered here.

The grid size we usually use in our simulation is $50 \times 50 \times 15$. To validate our numerical procedure, a series of calculations on the refined grids has been done. The spatial resolution has been doubled, while the time step has been decreased in two and four times. The comparison of integral properties of the solution, such as kinetic energy of the motion, Rayleigh number time history, flow structure, shows that grid refinement does not change the results significantly [13, 14].

6 Conclusions

Three-dimensional numerical simulation for LPE growth of ternary compounds confirm the existence of convective motion at Rayleigh numbers less than critical one predicted by linear stability analysis. The planform of subcritical convection and flow direction agree with theoretical data. The evolution of the flow pattern at Ra above the critical value is also consistent with theoretical predictions and experimental results. The numerical procedure is reliable and allows to perform long-time computer simulation of the convective motion in a wide range of operating parameters.

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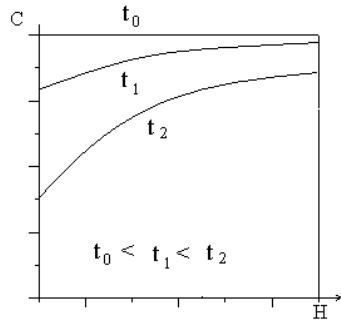


Figure 1: Concentration profile

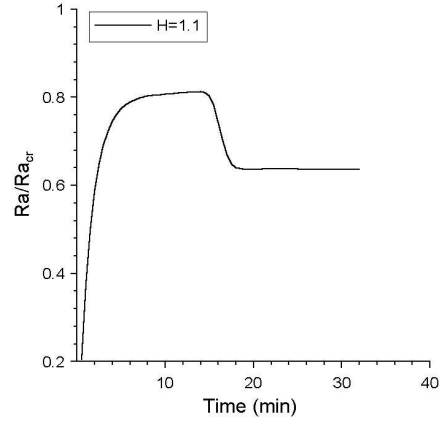


Figure 2: Time variation of Ra/Ra_{cr}

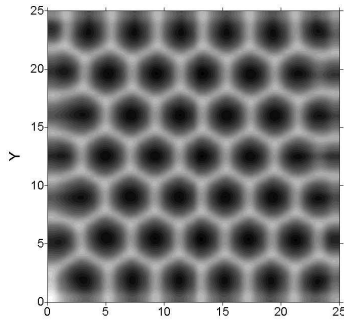


Figure 3: $Ra^x = 1 \cdot 10^3$

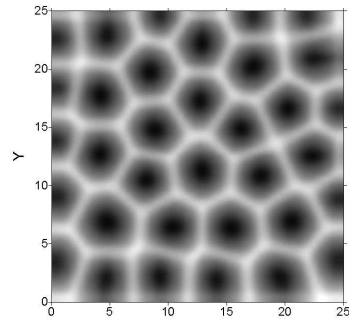


Figure 4: $Ra^x = 2.5 \cdot 10^3$

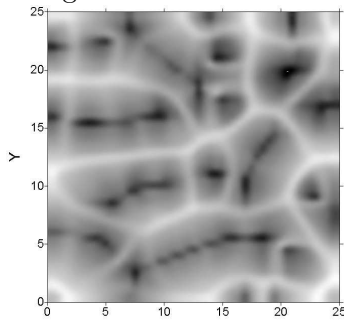


Figure 5: $Ra^x = 1.8 \cdot 10^4$

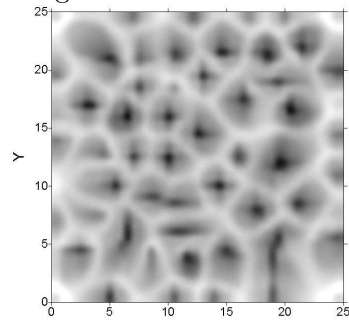


Figure 6: $Ra^x = 3.5 \cdot 10^4$

Concentration field distribution in plane $z = H$. Bright spots have higher concentration value than dark ones.