

### 3D COMPUTER SIMULATION OF TIME-DEPENDENT SOLUTAL CONVECTION.

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*Summary* The paper deals with computer simulation of natural convection in multicomponent solution. Three-dimensional calculations have been done to study the onset of convective motion and correspondent flow patterns. The process is considered for Rayleigh number in range  $1 \cdot 10^3 \div 4 \cdot 10^4$ . The results are in good agreement with theoretical data on hydrodynamics stability.

#### MATHEMATICAL MODEL

Computer simulation of mass-transfer in multicomponent systems with phase transition is one of the most challenging problems for modern crystal growth technology. The paper presents the numerical study 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 multi component semiconductor materials with desired structural composition. LPE is classified as a solution growth process. In distinction to solidification of pure materials or dilute alloys phase transition temperature in epitaxial growth depends on the composition of the liquid and solid phases.

The process described by 3D time-dependent fluid flow and mass transport equations. The solution is supposed to be incompressible and Boussinesq approximation is adopted. In cartesian coordinates  $(x, y, z)$  and nondimensional variables the set governing equations is:

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

where  $\partial_\xi \equiv \frac{\partial}{\partial \xi}$ ,  $\nabla = (\partial_x, \partial_y, \partial_z)$ ,  $\Delta = \nabla^2 = \partial_{xx}^2 + \partial_{yy}^2 + \partial_{zz}^2$ ,  $(x, y, z) \in \mathcal{D}$ ,  $\mathcal{D} = [0, L] \times [0, L] \times [0, H]$ ,  $L/H \approx 10$ ,  $V = (V_x, V_y, V_z)$  - velocity vector,  $p$  - pressure,  $C_i$ ,  $i = 1, 2$  - 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 Grashoff number for concentration,  $Sc_i = \nu / D_i$  is 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 – with  $H^2/\nu$  and concentration – 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:

$$V_{gr} = \frac{\partial_n C_1}{Sc_1(C_1^s - C_1)} = \frac{\partial_n C_2}{Sc_2(C_2^s - C_2)}$$

and phase diagram representing the equilibrium between the solution and growing layer:  $F_{liq}(C, T) = 0$ ,  $F_{sol}(C^s, C, T) = 0$ , where  $T(t)$  - temperature of the system, uniform in space and changing in time according a prescribed rule,  $C_i^s$  - concentration of the correspondent solute specie in the solid phase. On the rest of the boundary  $\partial_n C = 0$ . Boundary condition on velocity field is  $V = 0$ .

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. If the solution is slightly supersaturated dissolved components precipitate out of the solution onto the substrate. Supersaturation is usually maintained by gradually lowering the temperature during the growth. The reduction in the concentration of growth units in the vicinity of the growing layer gives rise to concentration gradients.

The onset of convection in LPE system has a known so far analogy. Neglecting the existence of the component with smaller Grashoff number and supposing linearity of phase diagram we actually obtain the problem concerning convective instability in a horizontal fluid layer with nonlinear conduction temperature profile. In our case instead of temperature the role of active scalar plays concentration. Theoretical analysis of the problem shows, that nonlinear profile gives rise to stable finite amplitude subcritical convective motion in the form of hexagons with flow direction at the center of the cells depending on the profile curvature sign.

## NUMERICAL RESULTS

The governing equations discretized at staggered grid using control-volume method. Approximation of convective terms ensure kinetic energy conservation and mass balance for dissolved components. The scheme is implicit, has second order in space, 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 follow predictor -corrector procedure. The calculated velocity field is substituted into the mass transfer equations, that are solved using the extended to 3D case coupled algorithm.

Full scale computer simulation for LPE growth of actual materials under reasonable operating conditions has been done for Rayleigh number  $Ra = \max_i Gr_i Sc_i$  varying in the range  $1.1 \cdot 10^3 < Ra < 1.1 \cdot 10^5$ ,  $Sc_i = 50$ .

The onset of finite amplitude subcritical convective motion is observed at  $Ra = 1100$ . The calculations show a transition from initial non-regular flow pattern to hexagonal planform. The run duration is approximately 25 vertical diffusion times. In the range  $1.1 \cdot 10^3 < Ra < 1.4 \cdot 10^4$  a regular cellular convection pattern is obtained. At Rayleigh number of  $1.4 \cdot 10^4 \div 3 \cdot 10^4$  the transition to skewed-varicose and knot instability is registered. 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. The mean cell size at  $Ra = 3.5 \cdot 10^4$  several times less then at  $Ra = 1.8 \cdot 10^4$ .

## CONCLUSIONS

Three-dimensional numerical simulation for LPE growth of ternary compounds confirm the existence of convective motion at Rayleigh numbers less than critical one measured in two-dimensional calculations and predicted by 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.

## References

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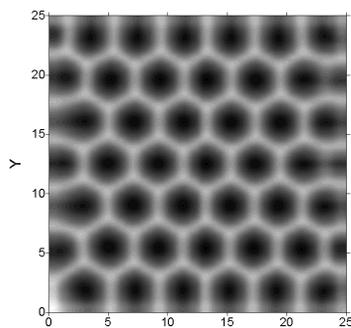


Figure 1.  $Ra^x = 1 \cdot 10^3$

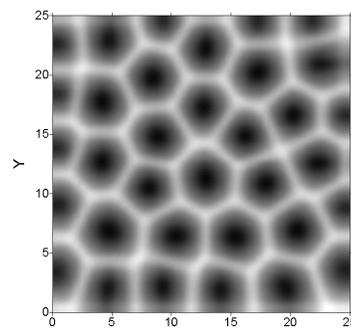


Figure 2.  $Ra^x = 2.5 \cdot 10^3$

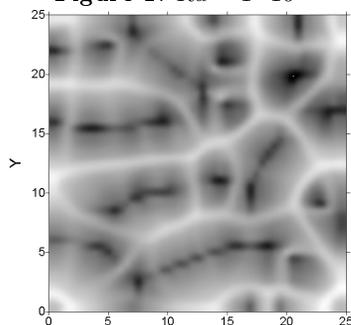


Figure 3.  $Ra^x = 1.8 \cdot 10^4$

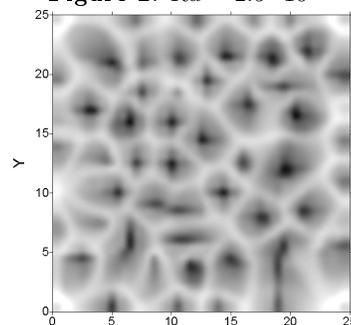


Figure 4.  $Ra^x = 3.5 \cdot 10^4$