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DOPANT DISTRIBUTION IN CRYSTALS GROWN BY THE SUBMERGED HEATER METHOD UNDER STEADY AND OSCILLATORY ROTATION

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ABSTRACT

The submerged heater method of crystal growth is simulated and the influence of steady and oscillatory rotation of crucible and/or heater on dopant distribution in crystals is investigated. Initial boundary value problem for the system of Navier-Stokes-Boussinesq equations is solved by the finite element method using the code ASTRA. The calculated history of dopant concentration at the solid-liquid interface is used to determine the dopant distribution in the grown crystals. © 1999 COSPAR. Published by Elsevier Science Ltd.

INTRODUCTION

The submerged heater method of crystal growth was introduced in Ostrogorsky et al (1990, 1995). An experimental study of the influence of rotation on dopant distribution in Ge-Ga crystals was carried out in Meyer et al (1997). The scheme of the method is shown in Fig. 1. The z-axis is the axis of axial symmetry, The submerged heater is moving slowly up and provides the desirable distribution of temperature inside the cylindrical crucible. The solid-liquid interface follows the heater so that the distance between the heater and the solid-liquid interface stays constant. The fat lines show the moving computational domain.

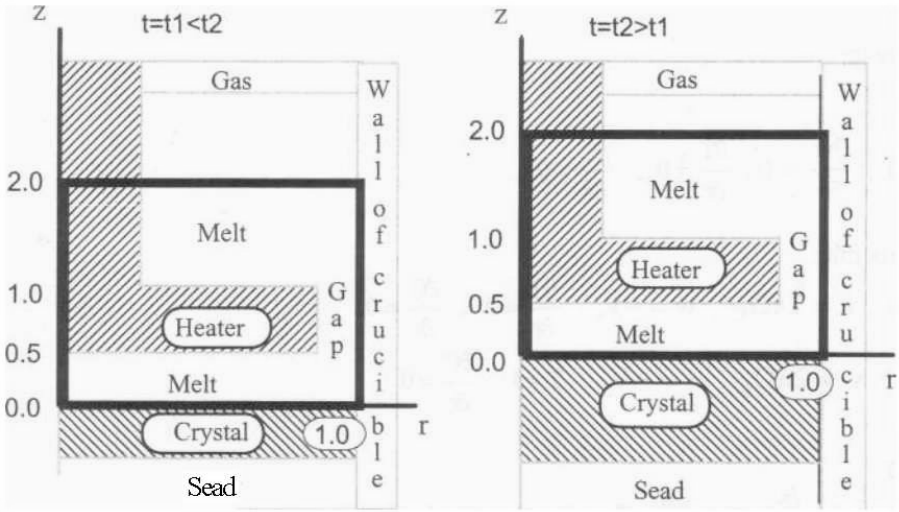

Fig. 1 Scheme of the Submerged Heater Method. The configuration is shown at two instants: tj and t2 > t], the fat lines show the moving computational domain.

The melt flows into the computational domain through the upper boundary, flows through the gap into the lower melt zone and solidifies at the solid-liquid interface (as if it flows out of the computational domain). Due to segregation of the dopant its amount in the melt is increasing. Convective flow of the melt and diffusion lead to a redistribution of the dopant in the melt and in the growing crystal. The goal of the present research is to study the influence of rotatory motion of the heater and/or crucible on the dopant distribution in growing crystals. In contrast to the mathematical study presented in Ostrogorsky (1990), Ostrogorsky and Dragojlovic (1995), and Meyer and Ostrogorsky (1997) the history of the process is taken into account here.

FORMULATION OF PROBLEM

It was assumed that the crystal growth process is axisymmetric, that the height of the work zone between heater and the solid-liquid interface as well as growth rate and thermal boundary conditions are constant, and that the solid-liquid interface is flat.

The system of Navier-Stokes-Boussinesq equations includes the continuity equation, momentum equations for r, θ, z directions, the heat transfer equation and the equation for the dopant concentration. The dimensionless equations read:

$$\frac{\partial u}{\partial r} + \frac{u}{r} + \frac{\partial w}{\partial z} = 0$$

$$\frac{\partial u}{\partial t} + u\frac{\partial u}{\partial r} + w\frac{\partial u}{\partial z} = -\frac{\partial p}{\partial r} + \mathrm{Pr}\left(\Delta u - \frac{u}{r^2}\right)$$

$$\frac{\partial v}{\partial t} + u\frac{\partial v}{\partial r} + w\frac{\partial v}{\partial z} + \frac{uv}{r} = \mathrm{Pr}\left(\Delta v - \frac{v}{r^2}\right)$$

$$\frac{\partial w}{\partial t} + u\frac{\partial w}{\partial r} + w\frac{\partial w}{\partial z} = -\frac{\partial p}{\partial z} + \mathrm{Pr}\,\Delta w - \mathrm{Pr}^2\left(\mathrm{Gr}\cdot T + \mathrm{Gr}_C \cdot C\right)$$

$$\frac{\partial T}{\partial t} + u\frac{\partial T}{\partial r} + w\frac{\partial T}{\partial z} = \Delta T$$

$$\frac{\partial C}{\partial t} + u\frac{\partial C}{\partial r} + w\frac{\partial C}{\partial z} = \frac{\mathrm{Pr}}{\mathrm{Sc}}\Delta C$$

where u, v, w - velocities, T - temperature, C - dopant concentration, p - pressure, Gr - Grashof number, Gr_C - solutal Grashof number, Pr - Prandtl number, Sc - Schmidt number. The nondimensional similarity numbers are used with their standard definition.

Boundary conditions read:

(i) axis of symmetry:

$$r = 0: \quad u = 0, \quad v = 0, \quad \frac{\partial w}{\partial r} = 0, \quad \frac{\partial T}{\partial r} = 0, \quad \frac{\partial C}{\partial r} = 0$$

(ii) side wall of the crucible:

$$r = 1,\ 0 \le z < 1: \quad u = 0, \quad v = 2\pi\Omega_C, \quad w = -V_s, \quad \frac{\partial T}{\partial r} = 0, \quad \frac{\partial C}{\partial r} = 0$$

$$r = 1,\ 1 \le z \le 2: \quad u = 0, \quad v = 2\pi\Omega_C, \quad w = -V_s, \quad T = 1, \quad \frac{\partial C}{\partial r} = 0$$

(iii) top inlet boundary:

$$0.25 \le r \le 1,\ z = 2: \quad u = 0, \quad \frac{\partial v}{\partial z} = 0, \quad \frac{\partial w}{\partial z} = 0, \quad T = 1, \quad C = 1$$

(iv) surface of heater:

$$0.0 < r < 0.25,\ z = 2.0:\ u = 0\ ,\ \ v = 2\pi\Omega_H^*\ ,\ \ w = 0\ ,\ \ T = 1$$

$$r = 0.25\ ,\ 1.0 < z < 2.0:\ \ u = 0\ ,\ \ v = 2\pi\Omega_H^*\ ,\ \ w = 0\ ,\ \ \frac{\partial C}{\partial r} = 0$$

$$0.25 < r < 0.985,\ z = 1.0:\ u = 0\ ,\ \ v = 2\pi\Omega_H^*\ ,\ \ w = 0\ ,\ \ \frac{\partial C}{\partial z} = 0$$

$$r = 0.985,\ 0.5 < z < 1.0\ :\ u = 0\ ,\ \ v = 2\pi\Omega_H^*\ ,\ \ w = 0\ ,\ \frac{\partial C}{\partial r} = 0$$

$$0.0 < r < 0.985,\ z = 0.5:\ u = 0\ ,\ \ v = 2\pi\Omega_H^*\ ,\ \ w = 0\ ,\ \ T = 0.7 + \frac{0.3r}{0.985}\ ,\ \ \frac{\partial C}{\partial z} = 0$$

(v) the solid-liquid interface:

$$z = 0:\ u = 0,\ v = 2\pi\Omega_C\ ,\ w = -V_S,\ T = 0\ ,\ \frac{\mathrm{Pr}}{Sc}\frac{\partial C}{\partial z} = V_S C(1-k)$$

where V_s - growth rate, $\Omega^*_H = \Omega_H \cos(2\pi\Omega_0 t)$ - frequency of submerged heater rotation , Ω_C - frequency of the crucible rotation, Ω_0 - frequency of oscillations, κ - segregation coefficient, C - concentration of dopant in the melt. The dopant concentration in the crystal C_{CR} is defined by the segregation ratio (Ostrogorsky (1990)):

$$C_{CR} = kC \quad (z = 0,\ 0 \le r \le 1)$$

The initial conditions read:

$$t = 0\ :\ u = 0\ ,\ v = 0\ ,\ w = 0\ ,\ T = 0\ ,\ C = 0$$

NUMERICAL METHOD

The finite element code ASTRA (Bourago (1988,1994)) was used to solve the problem. For a typical convection-diffusion equation

$$\frac{\partial A}{\partial t} + \mathbf{u}\cdot\nabla A = \nabla\cdot(\mu\nabla A) + F$$

the following implicit finite difference scheme in time was implemented:

$$\int_V \left(\frac{A^{n+1} - A^n}{\Delta t^n} + \mathbf{u}^n\cdot\nabla A^{n+1}\right)\left(\delta A + \Delta t^n \mathbf{u}^n\cdot\nabla\delta A\right)dV +$$

$$+\int_V \left(\mu_1^n \nabla A^{n+1}\cdot\nabla\delta A + F^n\delta A\right)dV = \int_S \mu_1^n \mathbf{n}\cdot\nabla A^{n+1}\delta A\,dV$$

Here the correction of the viscosity terms is used to provide a monotonous behavior of the solution:

$$\mu_1^n = \mu^n \frac{\mu^n}{\mu^n + 0.5\max(|\mathbf{u}^n\cdot\Delta\mathbf{x}|\,.\,|\mathbf{u}\Delta t^n|)}$$

Linear finite elements in space were used. Auxiliary algebraic problems were solved by the non-matrix conjugate gradient method with preconditioning by using the diagonal approximation of the stiffness matrix. The algorithm is unconditionally stable but for good accuracy the time step should approximately correspond to the Courant time

step $\Delta t_C = \min(|\Delta \mathbf{x}_i| / |\mathbf{u}_i^n|)$: $0.1\Delta t_C^n < \Delta t^n < 10\Delta t_C^n$. The incompressibility condition was fulfilled by the penalty method (1st algorithm), by the pressure correction Poisson equation (2nd algorithm), by using a stream function - vorticity formulation (3rd algorithm) and by Chorin artificial compressibility (4th algorithm).

Principal features of techniques mentioned above described in Peyret and Taylor (1990) where more references can be found.

Results for all used techniques are close enough to each other. Verification of the code ASTRA was made by solving known convection-diffusion test problems of Wheeler (1990) and de Vahl Davis and Jones (1983). Also the comparison with Ostrogorsky and Dragoilovic (1995) for steady state of submerged heater crystal growth was made and very good agreement of the results was found.

In order to simulate one case of submerged heater crystal growth it needs to calculate about $10^5 - 10^6$ time steps. Each run takes about 20 hours of CPU time on a IBM PC Pentium 200Mhz.

RESULTS

The case of Germanium melt and Gallium dopant was under study. The dimentionless input parameters and similarity numbers had the following values: Pr=0.0077, $\mathrm{Pr}^2 Gr = 300$, $\mathrm{Pr}^2 Gr_C = 0$, $\mathrm{Pr} \cdot Sc^{-1} = 0.0011$, V_S=0.0025, k=0.087. These values correspond to the case when the radius of the crucible is 16 mm, the width of the gap is 0.24 mm, the rate of crystal growth is 1.0 cm/hour, the frequency Ω_C is 18 rpm (dimensionless value is 2.0). Parametric calculations were performed for the set of runs shown in Table 1. The last column in Table 1 contains the calculated value of the radial inhomogeneity of the dopant distribution at the solid-liquid interface at the final instant: $\Delta C = C_{max} - C_{min}$.

The calculated distribution of the dopant in the crystals is presented in Fig. 2 for all six cases in Table 1: absence of rotation (R01), rotation of heater (R05), oscillatory rotation of heater (R12), rotation of crucible (R08), rotation of heater and crucible (R10,R11).

In most cases the dopant distribution becomes more homogeneous due to rotatory flow of the melt . Only the case of oscillatory rotation of heater shows a negative result.

Rotation can lead to a better mixing of the dopant and may level the dopant distribution at the solid-liquid interface. The rotation accelerates the vortex flow in the subdomain above heater and decreases natural convection in the subdomain under heater near the solid-liquid interface. Even in the case of steady rotation the flow is slightly oscillating in the subdomain under the heater. The traces of such oscillations can be seen in the dopant distribution in the crystal grown in the case of oscillatory rotation of the heater (R12).

A well known drawback of the non-rotating mode of crystal growth in a terrestrial environment is the existance of the peak of concentration in the crystal near the side wall of crucible under the gap. The rotation can help in the fight against this drawback. Our calculations indicate that the most homogeneous radial distribution of the dopant in the crystal may be reached due to simultaneous opposite rotation of crucible and heater (R10) .

Table 1. Script of runs

Run	$2\pi\Omega_H$	$2\pi\Omega_C$	$2\pi\Omega_0$	ΔC
R01	0.00	0.00	0.0	0.168
R05	2.00	0.00	0.0	0.102
R12	2.00	0.00	0.07	0.178
R08	0.00	2.00	0.0	0.061
R10	-0.33	2.00	0.0	0.049
R11	-2.00	2.00	0.0	0.054

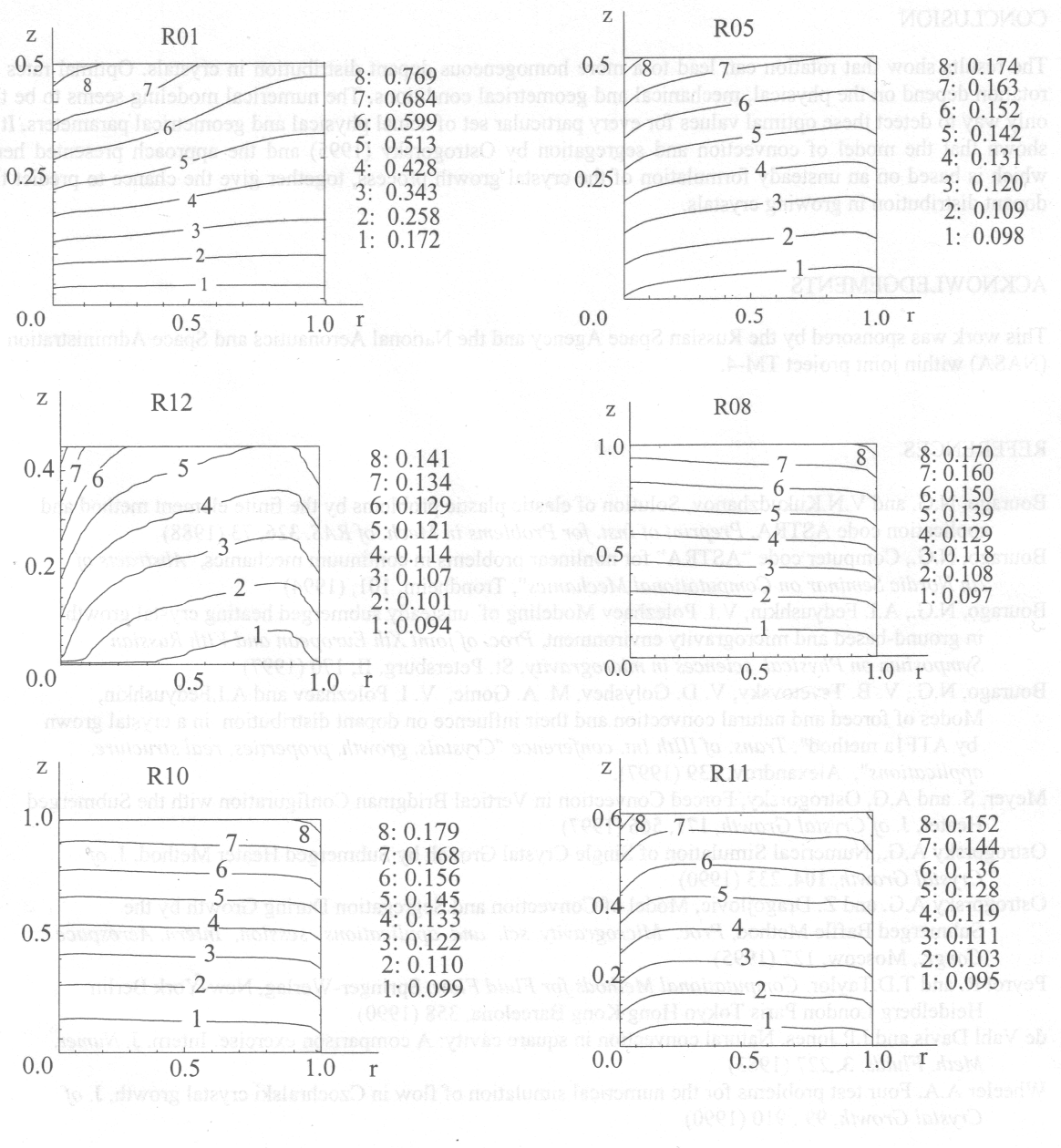

Fig. 2 The influence of rotation of heater and/or crucible on dopant distribution in the crystal.

For a microgravity environment (Gr=0) the effect of swirl melt flow was also studied and no big difference to the case of a terrestrial environment was found. It is understandable if the influence of rotation on the flow of the melt is much stronger than the influence of thermal buoyancy forces (if the rotation is fast enough like in our case).

During this research a great number of calculations were made to investigate also the influence of other input parameters such as geometrical parameters, thermal conditions, growth rate, initial dopant distribution, axial magnetic field, solutal buoyancy forces, properties of melt and dopant, vertical vibration of heater and/or crucible. Also the comparison of dopant distributions in crystals grown in a vertical Bridgman crucible with and without the heater/baffle was carried out. Some of these results were presented in Bourago et al (1997a, 1997b).

CONCLUSION

The results show that rotation can lead to a more homogeneous dopant distribution in crystals. Optimal rates of rotation depend on the physical, mechanical and geometrical conditions. The numerical modeling seems to be the only way to detect these optimal values for every particular set of actual physical and geometrical parameters. It is shown that the model of convection and segregation by Ostrogorsky (1995) and the approach presented here, which is based on an unsteady formulation of the crystal growth process, together give the chance to predict the dopant distribution in growing crystals.

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