THERMOCAPILLARY CONVECTION IN FLOATING-ZONE MELTING

I. V. Belova and G. B. Volkova UDC 532.5 + 536.421

The method of floating-zone melting is used to obtain high-quality single crystals or to remove impurities from a crystal. A liquid zone is formed in a cylindrical sample by a heater and is kept between the solid parts of the specimen by the surface tension of the melt. The temperature gradient at the free surface of the liquid phase changes the surface tension and this in turn causes thermocapillary convection of the melt. Vigorous studies are under way on mathematical problems that model floating-zone melting. Hydrodynamic, thermal and diffusion processes in the sample are considered separately, for the most part.

Numerical methods make it possible to analyze the solutions according to the characteristic parameters of the problem, the size of the samples, the thermal parameters of the heaters, etc. Numerical analysis, therefore, provides the necessary material for optimization of the process conditions. The objective of [1-3] was to ascertain how the size of the molten zone depends on the power of the heat source. The convection of the melt for a given temperature distribution on the lateral surface of the sample was studied in [4, 5], In [4] the front was assumed to be flat and given and in [5] the process was considered with the crystallization front in a position not known beforehand.

In this paper we study the effect of thermocapillary convection of the melt under weightlessness on the position and shape of the front under real boundary conditions on the lateral surface of the sample. Only the temperature distribution on the inner surface of the heater is usually given by the experimental data and so the mathematical model for calculating the heat and mass transfer has nonlinear boundary conditions of the third kind on the lateral surface (Fig. 1, where 1 is the heater, 2 is a gap, and 3 is the sample). The solution is assumed to be symmetric about the median plane of the molten zone, which is based on the results of [3, 4] as well as the proximity of the free surface of the melt to the cylindrical surface, which is true if the heat source is no wider than one diameter of the sample. Anisyutin [3] showed that with this condition the curvature of the free surface of the melt has very little effect on the shape of the crystallization front.

1. Formulation and Method of Solution of the Problem. In the cylindrical region $\{(r, z): 0 \le r \le 1, 0 \le z \le B\}$ (r and z are dimensionless coordinates, related to the radius R_1 of the sample, B is half the dimensionless length of the sample) we solve the stationary equation

$$\Delta U + \mathrm{Pr}\,\mathbf{V}\cdot(\nabla U) = 0. \tag{1.1}$$

Here U is the reduced temperature, related to the real temperature T by

$$U = (T/T_* - 1)/\alpha, \quad \alpha = \begin{cases} \varkappa_s/\varkappa_L, & T > T_*, \\ 1, & T \leqslant T_*; \end{cases}$$

where T_* is the crystallization temperature; $\mathrm{Pr} = \nu/\chi$ is the Prandtl number; and **V** is the vorticity. Then the position of the crystallization front is determined by the level line U = 0. The temperature distribution is easily found if the distribution of U is obtained.

The boundary conditions are:

at the symmetry axes

$$\partial U/\partial r|_{r=0} = \partial U/\partial z|_{z=0} = 0; \tag{1.2}$$

Novosibirsk. Translated from Prikladnaya Mekhanika i Tekhnicheskaya Fizika, No. 6, pp. 72-75, November-December, 1993. Original article submitted December 4, 1992; revision submitted January 11, 1993.

0021-8944/93/3406-0809$12.50

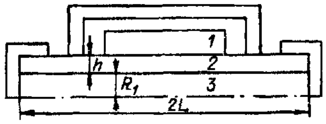

Fig. 1

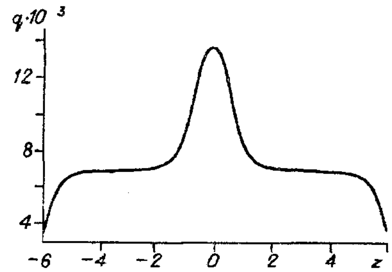

Fig. 2

at the end for z = B

$$U = T_1/T_* - 1 \tag{1.3}$$

(T_1 is the given temperature); at the lateral surface

$$\partial U/\partial n|_{r=1} = -Q\varepsilon\,\{(1 + \alpha U)^4/\alpha - q/\alpha\}, \tag{1.4}$$

where $Q = R_1\sigma T_*^3/\varkappa_L$ is a dimensionless quantity; ε is the blackness (depends on the temperature); σ is the Stefan–Boltzmann constant; and q is heat flux supplied. Figure 2 shows the distribution of q, calculated from the temperature distribution in the heater, obtained from the experimental data. The formula for calculating the heat flux (we ignore the contribution of reflected fluxes to the resultant flux) has the form [6]

$$q(z) = \sigma\varepsilon_2 R_1 \int_{-L}^{L} \frac{h^2 T_2^4}{(h^2 + (z - s)^2)^2}\, ds.$$

Here ε_2 and T_2 are the blackness and the temperature distribution at the heater wall; and h is the gap between the surface of the sample and that of the heater.

Equation (1.1) with boundary conditions (1.2)-(1.4) was solved by the method of finite elements, with the region divided into eight-node isoparametric elements [7].

The vorticity components are the solution of the following problem: $\mathbf{V} \equiv \mathbf{0}$ for $T \le T_*$. For $T > T_*$ in the region $0 \le r \le 1$, $0 \le z < f(r)$ ($z = f(r)$ is the position of the crystallization front) we solve the system of equations

$$u = -\frac{1}{r}\frac{\partial\psi}{\partial z}, \quad v = \frac{1}{r}\frac{\partial\psi}{\partial r}, \quad \omega = \frac{\partial v}{\partial r} - \frac{\partial u}{\partial z}$$

(u, v are the vorticity components, ψ is the stream function, ω is the nonzero current vector component),

$$u\frac{\partial\omega}{\partial r} + v\frac{\partial\omega}{\partial z} - \frac{u\omega}{r} = \frac{\partial^2\omega}{\partial r^2} + \frac{1}{r}\frac{\partial\omega}{\partial r} + \frac{\partial^2\omega}{\partial z^2} - \frac{\omega}{r^2},$$

$$\frac{\partial^2\psi}{\partial r^2} - \frac{1}{r}\frac{\partial\psi}{\partial r} + \frac{\partial^2\psi}{\partial z^2} = \omega r$$

with the boundary conditions

$$\psi|_{r=0} = \psi|_{r=1} = \psi|_{z=0} = \psi|_{z=f(r)} = 0, \quad \left.\frac{\partial\psi}{\partial n}\right|_{z=f(r)} = 0,$$

$$\omega|_{r=0} = \omega|_{z=0} = 0, \quad \omega|_{r=1} = \frac{\sigma_T \alpha T_* R_1}{\rho\nu^2}\frac{\partial U}{\partial z}.$$

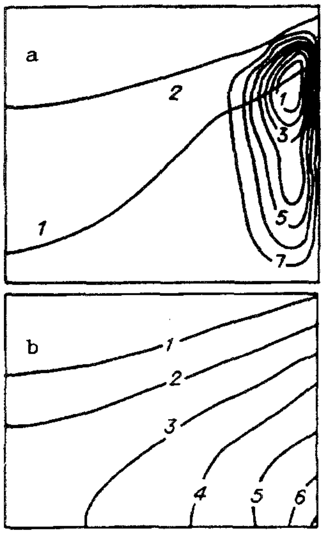

Fig. 3

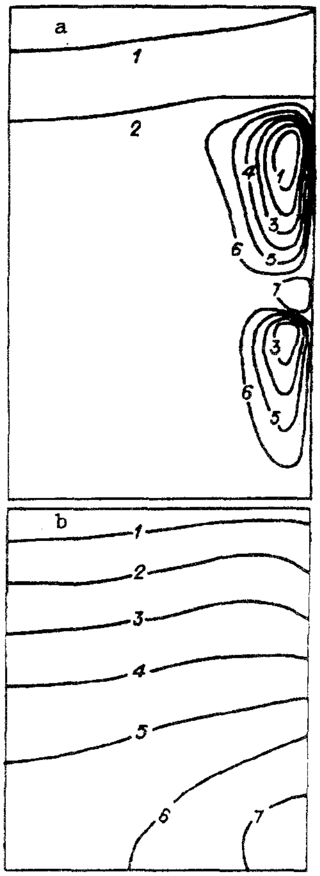

Fig. 4

The problem for the current on the solid wall, the interface, was completed by using the method of direct satisfaction of the boundary conditions, described in [8]: in this method the difference analog of the boundary condition of zero slip $(\partial\psi/\partial n)_b = 0$ is satisfied in each transient layer directly, this being achieved by adjusting the stream function ψ near the boundary.

The determining criteria in the problem are the Prandtl number, the vortex strength on the free surface, the dimensionless heat flux $\bar{q} = Q\varepsilon q/\alpha$, and the reduced temperature at the boundary $z = B$.

The equations of hydrodynamics in the variables of the stream-vortex function are solved by the method of fixing. Stationary equations are replaced by nonstationary equations with the introduction of an iteration parameter analogous to time. A scheme of variable directions is then used [8]. The procedure described in [9] for determining the optimum parameters is used to optimize the calculations. In this procedure, the estimates of the maximum and minimum eigenvalues of the problems are adjusted on the basis the results from the previous steps.

The problem (1.1)-(1.4) was solved together with the hydrodynamic equations as follows: We assumed that initially $\mathbf{V} = \mathbf{0}$. The shape of the front (without convection) was determined and then the convection was calculated. The velocities so found were inserted into (1.1) and the new shape of the front was determined. The iteration process was continued until the shape of the front was fixed.

2. Results of Calculation. The calculations were made with experimental data obtained with a sample of dimensions $R_1 = 0.79$ cm, $L = 4.8$ cm, $h = 0.56$ cm. The characteristics of the material correspond to germanium. The results of the calculations for $T_1 = 925$ and 949 K are shown in Figs. 3 and 4, respectively. Line 1 in Figs. 3a and 4a corresponds to the position of the front obtained in calculations without convection of the melt and line 2 corresponds to the position of the front with convection in the melt. Figures 3a and 4a also show the level lines of the stream function and Figs. 3b and 4b, the isotherms in the liquid phase. In Figs. 3a and 4a we see two vortices, in the same direction and separated by a small region of almost total quiescence. The vortices are localized along the free boundary of the melt, because of the boundary layer in this region and the absence of other mechanisms that could produce convection. The calculations suggest that thermocapillary convection substantially affects both the position and shape of the interface. On the whole, the front becomes less curved, but a slight curvature near the lateral surface is caused by the local effect of thermocapillary convection. This convection may make the molten zone either wider or narrower, depending on the thermal conditions at the ends of the preform (Figs. 3a and 4a). On the basis of the data [7] and the information obtained about the effect of convection on floating-zone melting, we can say that by smoothing our the interface thermocapillary convection creates a stressed state in the growing solid phase that is more favorable for technical purposes. Regions in which the shear stress intensity exceeds the critical value and results in a substantially higher dislocation density may not form at all in this case or if they do such regions are localized along the lateral surface of the sample. The change in the width of the molten zone must be taken into account, of course, since a critical value that determines the region of stability for the process exists for this process parameter.

We are indebted to V. V. Kuznetsov and O. M. Lavrent'eva for useful discussion of this work.

REFERENCES

1. D. K. Donald, "Thermal regime under the conditions of vacuum melting," Prib. Nauchn. Issled., No. 7 (1961).
2. N. Obayashi, "Power required to form a floating zone and the zone shape," J. Crystal Growth, **43,** 417 (1978).
3. B. M. Anisyutin, "Numerical analysis of the heat problem for floating-zone melting," in: Problems of Fluid Mechanics and Heat and Mass Transfer with Free Boundaries [in Russian], Mezhvuz, Sb. Nauch. Tr., Novosib. Gos. Univ., Novosibirsk (1987).
4. S. G. Slavchev and Zh. D. Kozhurkharova, "Numerical simulation of thermocapillary convection in a noncylindrical floating zone of a crystal," in: Applications of Numerical Methods [in Russian], Sofia (1985).
5. Yu. V. Ananovich and E. D. Lyumkis, "Simulation of heat and mass transfer processes during floating-zone melting," Proceedings of Third All-Union Conference on Simulation of Crystal Growth [in Russian], Riga (1990).
6. M. Jacob, Heat Transfer, Vol., Wiley, New York (1957); Vol. II (1957).
7. I. V. Belova, "Calculation of the temperature and thermoelastic stresses in single crystals in the floating-zone technique," in: Mathematical Simulation. Applied Problems of Mathematical Physics [in Russian], Riga (1990).
8. V. I. Polozhaev, A. V. Buné, N. A. Berezub, et al., "Mathematical simulation of convective heat and mass transfer on the basis of the Navier–Stokes equations," Nauka, Moscow (1987).
9. L. A. Hageman and D. M. Young, Applied Interactive Methods, Academic Press, New York (1981).