D12

THERMOCAPILLARY FLOW IN A CYLINDRICAL LIQUID DROP AT ZERO GRAVITY*

Chong E. Chang and William R. Wilcox
Chemical Engineering and Materials Science Departments
University of Southern California, Los Angeles, California 90007

ABSTRACT

Surface tension driven flow in the floating zone melting geometry was studied by means of computer solution of the differential equations for heat and momentum transfer. At small temperature differences one cell is formed in the top of the zone, and its mirror image in the bottom of the zone. They appear somewhat like vortex rings (smoke rings). As the temperature differences increase, secondary cells form and the flow probably becomes oscillatory. High differences produce turbulence. For silicon, effective heat shields are required to avoid turbulence, and the flow is not appreciably influenced by zone motion or by earth's gravity. The laminar cells do not have a strong influence on the heat transfer with good thermal conductors. Radiant heating of poor thermal conductors may result in oscillatory flow even with small driving forces.

INTRODUCTION

Floating zone melting is used commercially and in research for crystal growth and for purification of high melting materials. The melt does not contact a crucible, as shown schematically in Fig. 1. Heating may be by radiation, by an electron beam, or by induction from a surrounding high frequency coil. The zone is moved through the solid either by moving the heater or by moving the solid rods bordering the zone. There are five sources of convection in a floating zone:

1) When the zone is taken as stationary, there is a flow through the zone generated by melting at one solid-liquid interface and freezing at the other, due to movement of the zone through the solid.

2) Convection due to rotation of the two solid rods. (Slow rotation is sometimes used to maintain a cylindrical zone and growing rod.)

3) With induction heating one obtains electromagnetic stirring, which is difficult to calculate.

4) Buoyancy-driven natural convection due to interaction of a gravitational or accelerational field with density variations in the melt.

5) Surface-tension driven flow due to variations in surface tension along the melt surface.

*Supported by NASA under contract NAS8-29847

The purpose of the calculations described here was to estimate the magnitude of the surface-driven (Marangoni) flow and its influence on heat and mass transfer. Mass transfer influences the degree of purification and the homogeneity of single crystals. Heat transfer influences the perfection of the crystals, the stability of the zone, and sometimes influences the homogeneity as well, through its effect on the freezing interface shape. In order to determine the influence of surface driven flow on zone melting, the partial differential equations for momentum and heat transfer were considered. The momentum equations were expressed in finite difference cylindrical stream-function form, and solved by the Gauss-Seidel iteration method (1). The Newton-Raphson method was used to solve the heat transfer equations, with an over-relaxation parameter of 1.9 for low Prandtl numbers and an underrelaxation for high Prandtl numbers. Solutions were obtained for two situations--a parabolic temperature profile along the melt-vapor surface and for a ring heat source at the center of the zone. The parabolic profile corresponds approximately to radiant heating, and the ring source corresponds to electron beam heating. The mass transfer equation coupled with the computed flow fields were employed to find the impurity concentration fields in the melt at steady state.

The results of the calculations are expressed in dimensionless form and for the physical properties of silicon with a 1 cm x 1 cm zone, as a concrete example. For molten silicon the melting point is 1410°C, the Prandtl number is 0.023, the Schmidt number is about 5, $\rho_f(\partial\gamma/\partial T)/\mu^2$ is 14,000, and the emmissivity is assumed to be 0.3 for both the melt and the solid. Earth's gravitational field and ordinary zone travel rates were found to have negligible influence on the convection for silicon.

RESULTS

Dimensional analysis of the momentum equations produced a parameter charactering the driving force for surface driven flow, $M = a(T_o-T_m)\rho_f(\partial\gamma/\partial T)/\mu^2$, where a is the radius of the zone, T_o is the temperature of the surface of the melt at the center of the zone, T_m is the interface temperature, ρ_f is the density of the melt, γ is the surface energy or surface temperature, and μ is the viscosity.

Streamlines for a Parabolic Temperature Profile at the Free Melt Surface (Radiation Heating)

For simplicity, a parabolic temperature profile along the melt surface was assumed for our initial calculations. This enabled us to solve the momentum equations without simultaneously solving the heat transfer equations. A parabolic profile, with a maximum at the center, seems reasonable for radiant heating. The dimensionless surface tension parameter M increases as the radius of the zone a and the temperature variation along the melt surface increases. In Figures 2 through 4, the streamlines for parabolic temperature profiles are shown to illustrate the effect of increased values of M (350, 3500 and 7000). Donut-shaped vortex cells were formed. With M = 35 and 350 only two cells are generated, and the centers of the vortices move closer toward the liquid/solid interfaces as M increases. With M = 3500, however, which corresponds to a condition of a = 0.5 cm and (T_o-T_m) = 0.5°C for silicon,* secondary vortex cells were induced behind the primary vortex, as shown in Figure 3. As the value of M was further increased up to 7000, third and fourth vortices were produced, as shown in Figure 4. These multiple vortices are probably indicative of oscillations and incipient turbulence, which cannot be calculated in a steady state analysis (oscillations are frequently found with free convection in enclosed cavities between laminar and fully turbulent flow). The maximum velocity of the melt for 1 cm diameter silicon with (T_o-T_m) = 1.0°C is calculated to be 2 cm/sec.

Fluid Flow Coupled with Heat Transfer
(Electron Beam Heating)

Electron beam heating is commonly used in floating zone melting and was considered as another heating mode. A ring heat source at the center of the zone was assumed. Rather than specify the power input, the circumferential temperature T_o at the center of the zone was specified for convenience in analysis. The procedure used to solve the coupled heat-momentum transport problem was as follows:

1) The temperature field for conduction was computed by neglecting convective heat transfer.

2) The surface temperature profile from 1) was utilized to calculate the first approximate solution of fluid flow.

3) The temperature field was recalculated using the flow fields from 2).

4) Steps 2) and 3) were repeated until the temperature and the fluid flow fields no longer changed appreciably.

The resulting streamlines with M = 350** are drawn in Figure 5 in which we took values for silicon with the surroundings at the melting point, i.e., the heat shielding about the zone is extremely effective. While this does not correspond exactly with reality, it does show the correct features. Comparing with Figure 2, we see that the vortex centers are shifted nearer to the heat source from the liquid/solid interfaces. This is because the steepest temperature gradient is at the center of the zone. The maximum velocity for silicon was 0.55 cm/sec. Comparing the temperature fields with surface tension driven flow for M = 350 with those for pure conduction, there was no significant change except for a slight one near the center of the zone. This indicates that conduction is much greater than convective heat transfer as would be expected for the small Prandtl number for silicon. The vorticity fields for M = 350 are shown in Figure 6. The maximum vorticity and its location for various conditions are also summarized in Table I.

Influence of Gravity on Flow

In a vertical silicon melt with M = 350 at earth's gravity the flow and

*Without heat shielding, we estimate the temperature difference (T_o-T_m) would be on the order of 10 to 20°C, and the flow would be turbulent.

**For electron beam heating of 1 cm diameter silicon, the Marangoni number Ma is about 1/50 of the value of M. Without heat shielding Ma is estimated to be about 60,000.

temperature fields in the floating zone do not change appreciably from those at zero g.* When the acceleration is increased to ten times the earth's gravity, the lower vortex contracts while the upper one is expanded, particularly at the center of the zone where the flow field is relatively weak. This is shown in Figure 7 and may be compared with Figure 5 at zero g. This means that surface driven flow predominates even on earth for many materials for radiant and electron beam heating. It may even be important when induction heating is employed and may account for some of the compositional inhomogeneities observed.

Influence of Prandtl Number

The Prandtl number was increased from 0.023 to 0.3 and 2.0 by increasing the specific heat, and keeping the viscosity and the thermal conductivity of the melt constant. As the Prandtl number increases, the temperature gradients along the free liquid surface (for fixed $T_O - T_m$) increase near the heat source and also near the liquid/solid interfaces, but decrease significantly in between. Since the convective heat transfer becomes more significant as a result of increasing the Prandtl number, the isotherms are increasingly distorted from those of pure conduction. This, in turn, causes changes in the flow field. For example, the center of the vortex cell near the liquid/solid interface shifts closer to the interface as the Prandtl number increases, as shown in Figure 8.

Influence of Zone Travel on Hydrodynamics

The effect of zone motion on the flow field was found to be negligible in all of our calculated streamlines for zone travel rates of up to 5 cm/hour in silicon.** However, the effect becomes significant when the zone travel rate becomes comparable with the velocity of surface-driven flow.

The flow field for electron beam heating with $M = 35$ is taken as a model case in order to show the influence of zone travel on hydrodynamics in the floating zone. The maximum velocity of the melt for $M = 35$ is 0.07 cm/sec (or 250 cm/hour), and the average velocity is about 70 cm/hour. In Figure 9, the streamlines in floating zone melting of silicon at zero g are shown for a freezing rate of 30 cm/hour. The lower vortex cell floats away from the bottom interface and its size is reduced as the zone travel rate increases.

CONCLUSION

For silicon we have seen that thermocapillary flow is very vigorous, and is turbulent when heat shields are not employed. The buoyancy flow is negligible in comparison. With moderate heat shielding, oscillatory convection is likely, but was not studied here. Zone motion does not have an appreciable effect on the convection unless the temperature gradients along the melt surface are made very small by effective heat shielding. With laminar flow, the convection has only a small influence on heat transfer at small Prandtl number ($Pr \ll 1$), but a large influence for $Pr > 1$.

---

*Only temperature variations were considered.

**A zone travel rate of 5 cm/hour is typical for growth of single crystals, while faster rates are employed for vacuum outgassing and lower rates for zone refining.

REFERENCES

1. B. W. Arden and K. N. Astill, "Numerical Algorithms: Origin and Applications," Addison-Wesley, Reading, Mass. (1970)

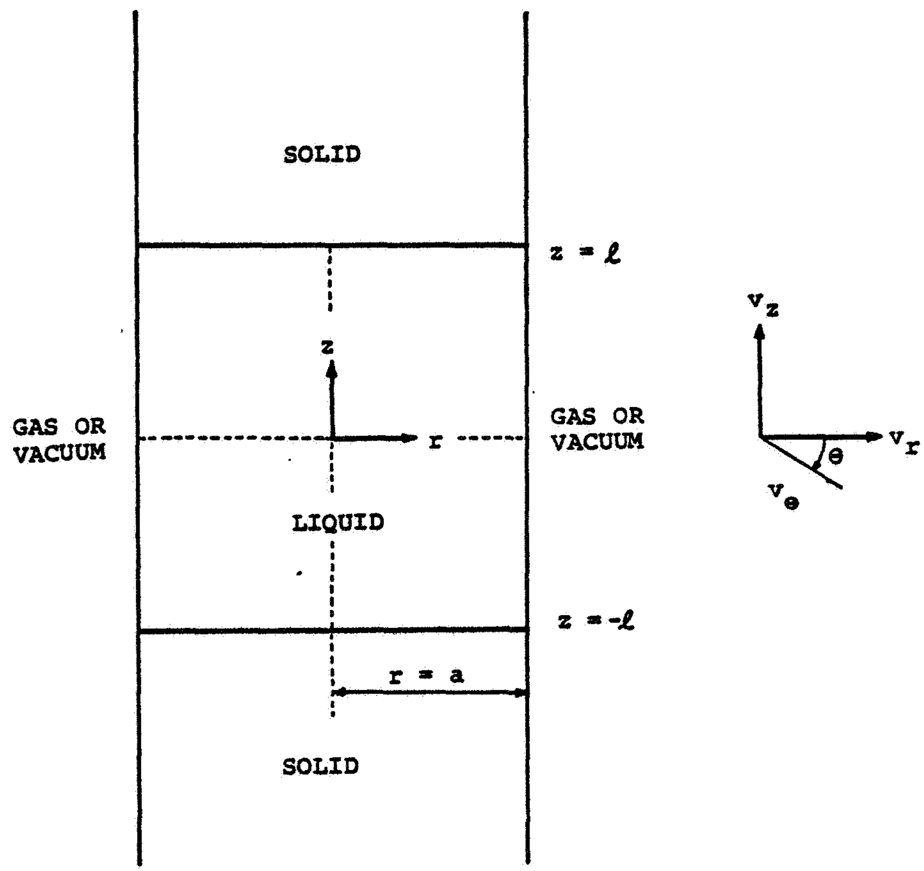

Fig. 1. Idealized diagram of floating zone melting.

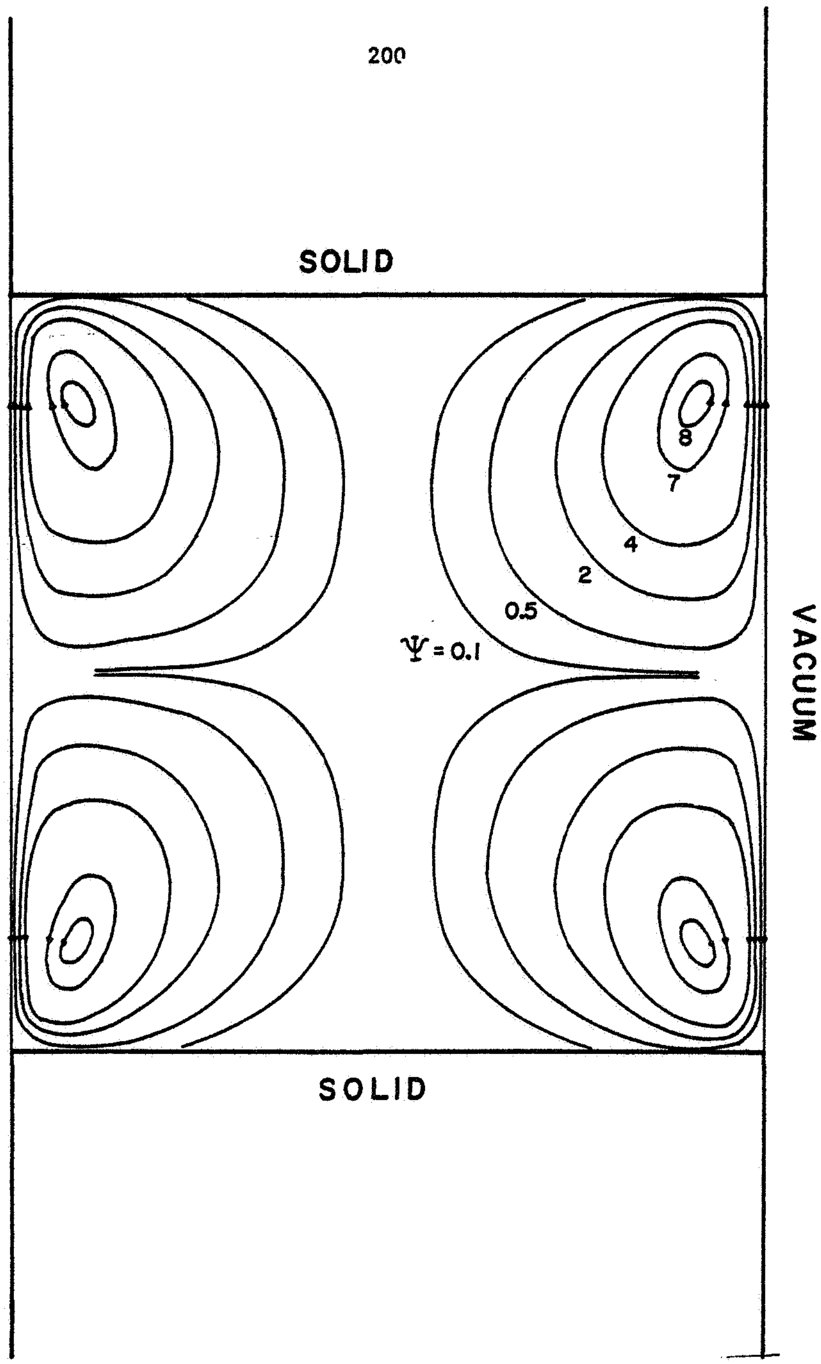

Fig. 2. Computed dimensionless streamlines ϕ for surface tension driven flow in a floating zone at zero gravity with a parabolic temperature profile on the free liquid surface with M = 350 and $v_c = 0$. For silicon with $(T_o-T_m) = 0.05^oC$ and a = 0.5 cm.

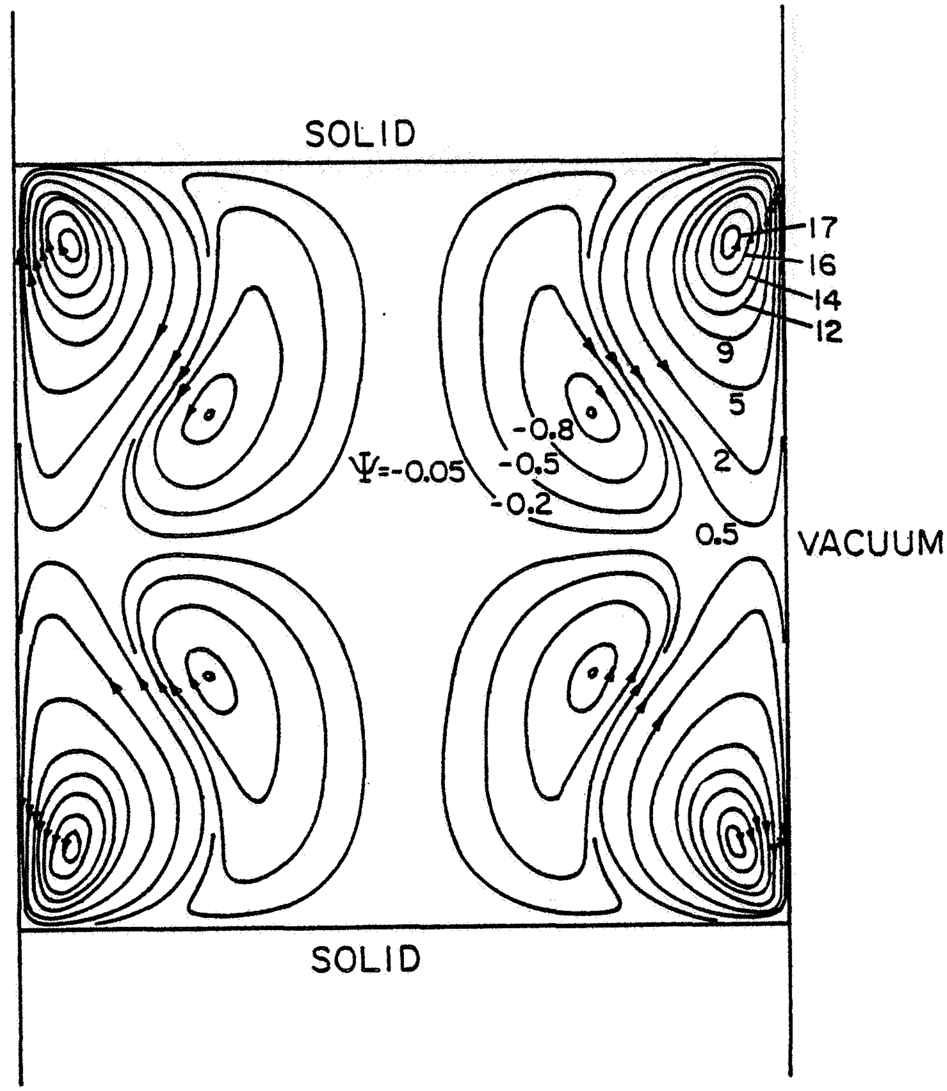

Fig. 3. Computed dimensionless streamlines ψ for surface driven flow in a floating zone at zero gravity with a parabolic temperature profile on the free liquid surface with $M = 3500$ and $v_c = 0$. Silicon with $(T_o-T_m) = 0.5^oC$ and $a = 0.5$ cm.

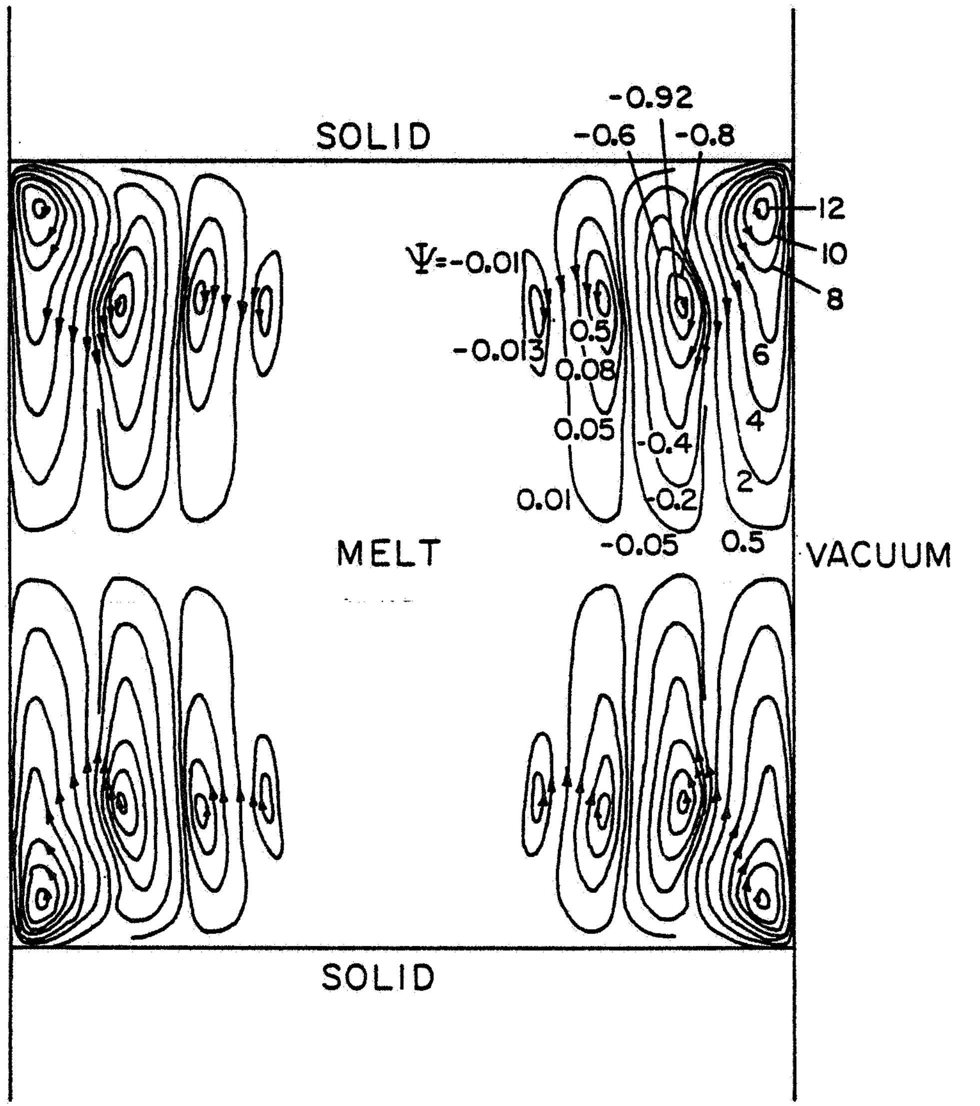

Fig. 4. Computed dimensionless streamlines ψ for surface tension flow in a floating zone at zero gravity with a parabolic temperature profile on the free liquid surface with M = 7000 and v_c = 0. Silicon with (T_o-T_m) = 1.0°C and a = 0.5 cm.

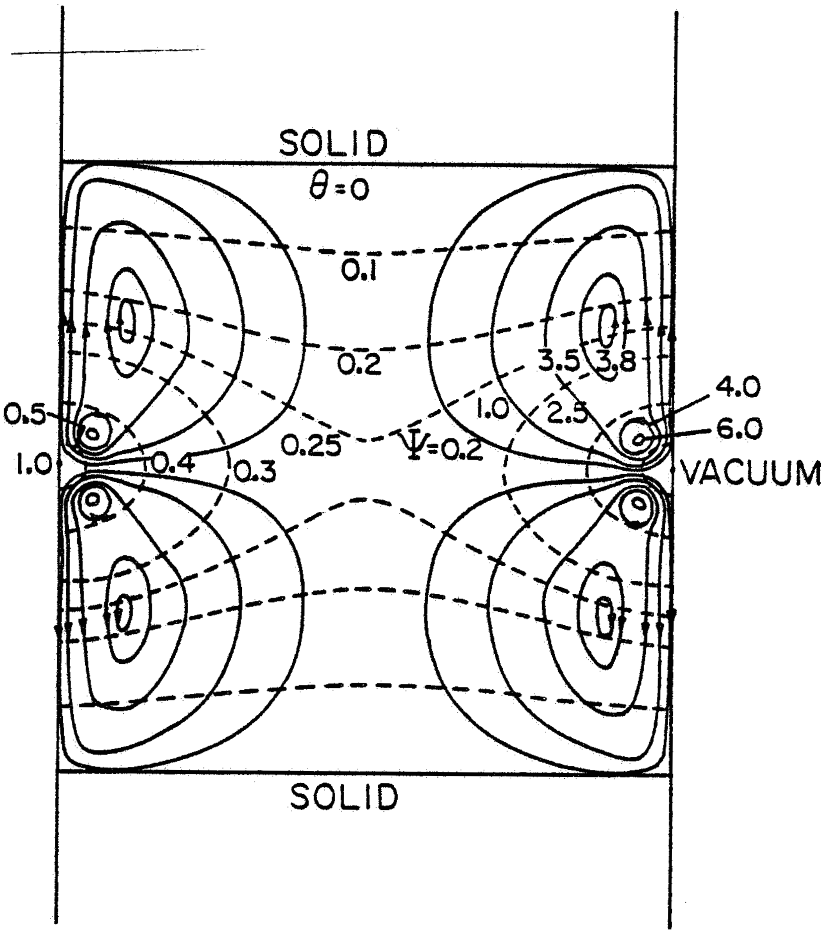

Fig. 5 Isotherms θ and streamlines ψ at zero gravity for electron beam heating of silicon with $(T_o-T_m) = 0.05°C$, $T_c = T_m$, $\varepsilon_s = 0.3$, $a = 0.5$ cm, $M = 350$, $Pr = 0.023$, $v_c = 0$.

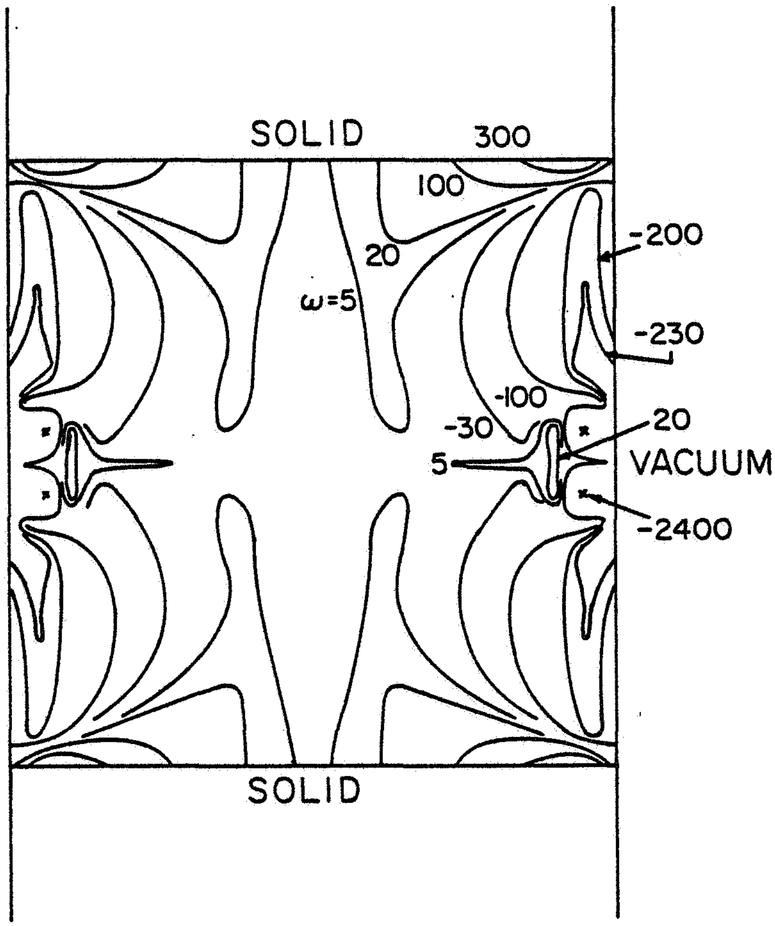

Fig. 6. Dimensionless vorticity field ω corresponding to the streamline field shown in Figure 4.

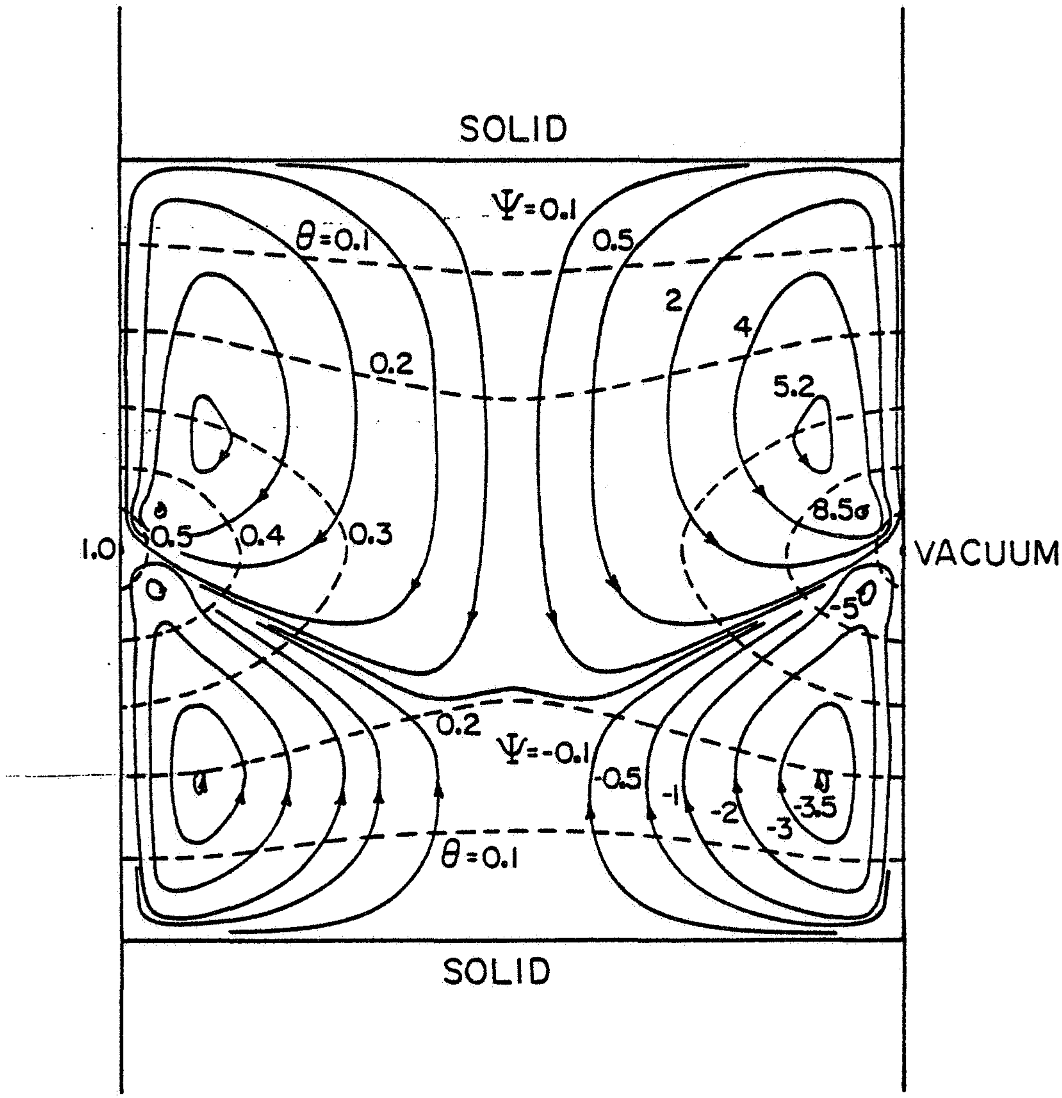

Fig. 7. Streamlines and isotherms for combined surface-driven and buoyancy-driven flow with Gr_h = 775. Other conditions are the same as Figure 4.

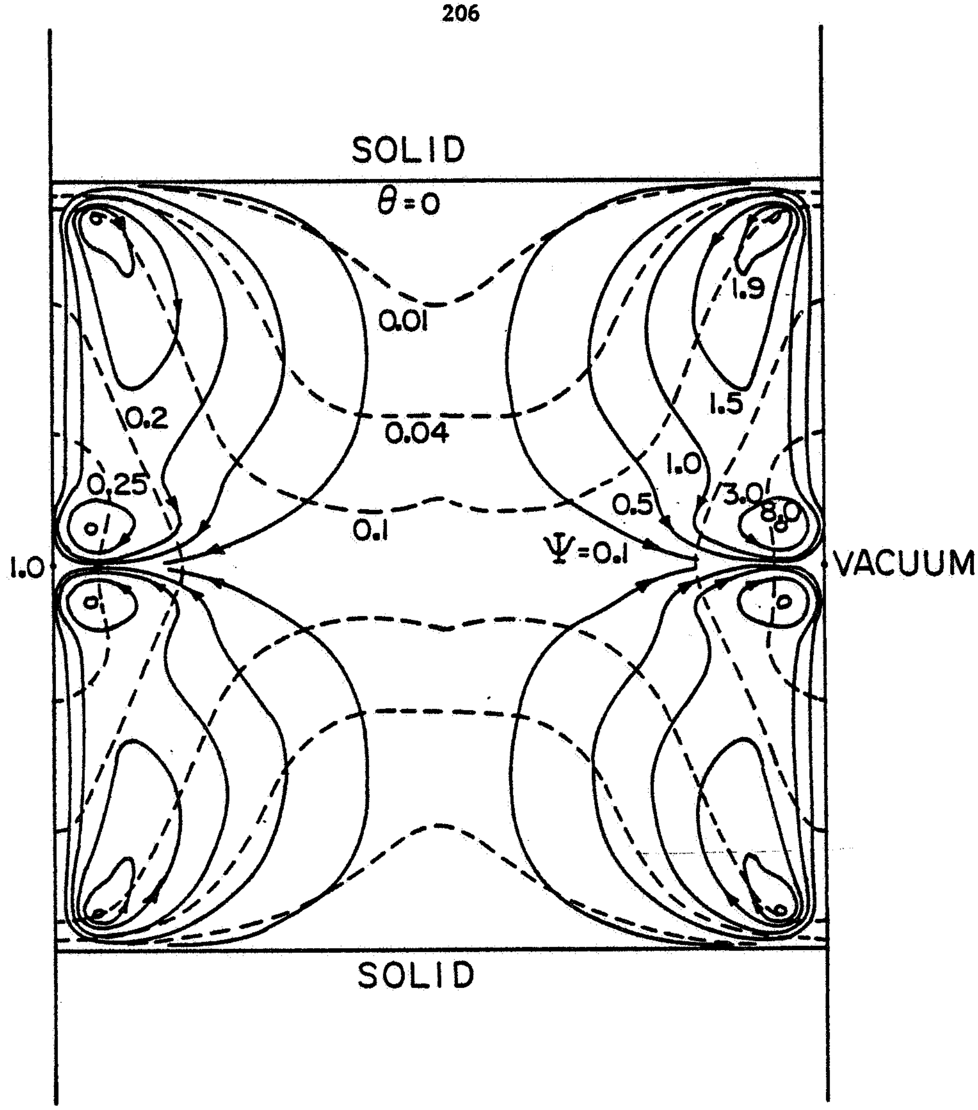

Fig. 8. Isotherms θ and streamlines ψ for the same conditions as in Figure 4, except Pr = 2.

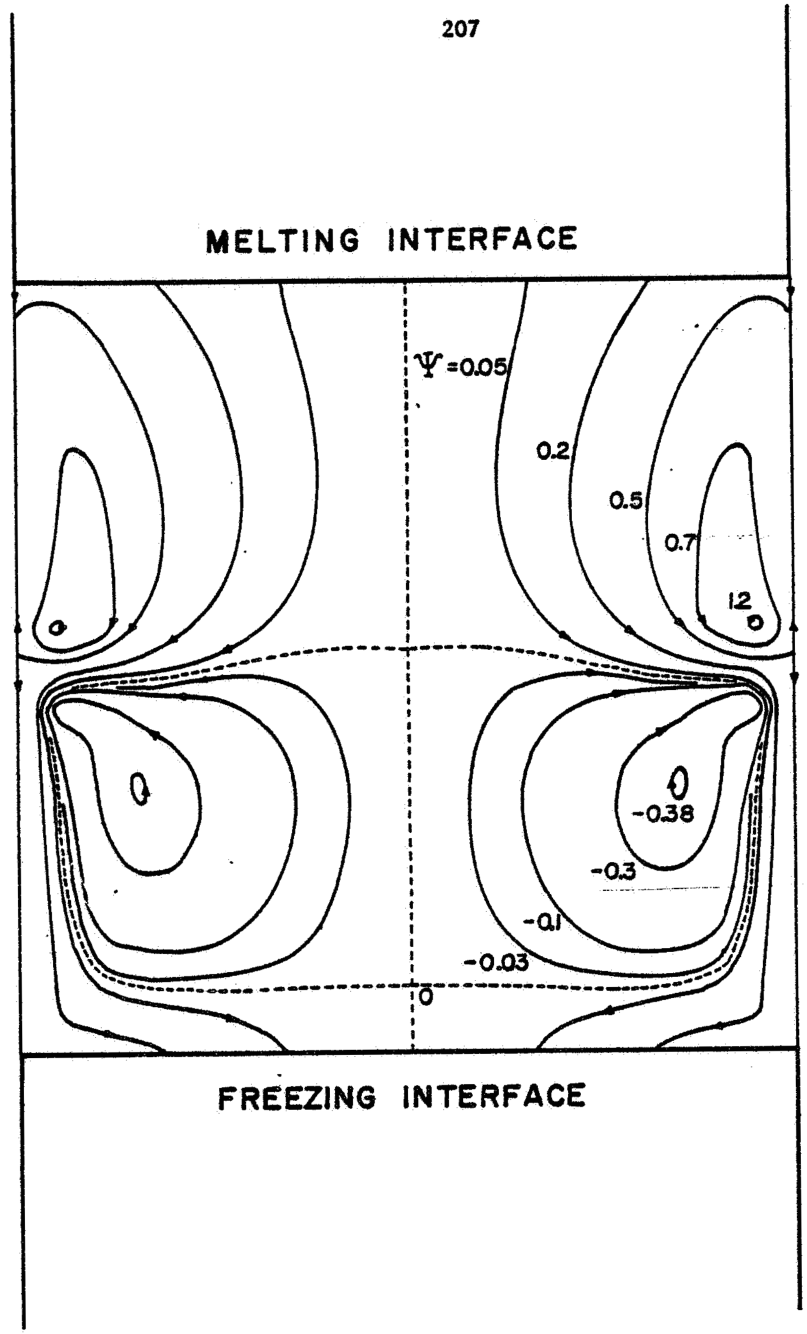

Fig. 9. Streamlines ψ at a zone travel rate of 30 cm/hour with M = 35, Pr = 0.023. For silicon, a = 0.5 cm, $(T_o - T_m)$ = 0.005°C, $T_c = T_m$, ε_s = 0.3.