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SPACE-BASED CRYSTAL GROWTH AND THERMOCAPILLARY FLOW

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Summary

The demand for larger size of crystal is ever increasing especially in applications associated with the electronic industry, where large and pure electronic crystals (notably silicon) are the essential material to make high-performance computer chips. Crystal growth under weightless conditions has been considered an ideal way to produce bigger and hopefully better crystals. One technique which may benefit from a microgravity environment is the float-zone crystal-growth process (fig. 1), a container-less method for producing high-quality electronic material. In this method, a rod of material to be refined is moved slowly through a heating device which melts a portion of it. Ideally, as the melt resolidifies it does so as a single crystal which is then used as substrate for building microelectronic devices. The possibility of contamination by contact with other material is reduced because of the "float" configuration. However, since the weight of the material contained in the zone is supported by the surface-tension force, the size of the resulting crystal is limited in Earth-based productions; in fact, some materials have properties which prevent this process from being used to manufacture crystals of reasonable size. Consequently, there has been a great deal of interest in exploiting the microgravity environment of space to grow larger size crystals of electronic material using the float-zone method.

In addition to allowing larger crystals to be grown, a microgravity environment would also significantly reduce the magnitude of convection induced by buoyancy forces during the melting stage. This type of convection was once thought to be at least partially responsible for the presence of undesirable nonuniformities in material properties called striations observed in float-zone material. However, past experiments on crystal growth under weightless conditions found that even with the absence of gravity, the float-zone method sometimes still results striations. It is believed that another mechanism is playing a dominant role in the microgravity environment.

Thermocapillary Convection

In float-zone crystal growth, the temperature difference between two ends of the zone produces a surface-tension gradient along the interface between the liquid and the surrounding gas causes a fluid motion in the melt. This type of flow is called thermocapillary convection, and will exist in any gravitational environment. Experiments on this type of convection have found that, under certain conditions, the flow is in a stable circulating state. When the conditions change, the flow also changes to an oscillating, unstable state. The condition is a combination of factors, including temperature differences, the material grown and the size of the crystal. Recent speculation is that the onset of time-dependent thermocapillary convection (ref. 5) is actually responsible for the appearance of the striations and nonuniformity in the final product. Since this mode of flow will exist in crystal-growth process, the stability properties of thermocapillary convection are of possible technological importance.

Experiments have been performed in half-models of the float-zone process, so termed because they are meant to simulate the lower half of a float-zone melt, where the axial buoyancy gradient is stabilizing, meaning the hotter fluid is on top of the colder fluid. In one of the experiments (ref. 1), a liquid bridge is established between a pair of cylindrical rods, which are differentially heated, with the upper rod held at a higher temperature than the lower. For small enough temperature differences, as characterized by a dimensionless parameter called Marangoni number $Ma = \gamma(T_H - T_C)R/\kappa\mu$ steady convection is observed, while for large values, a transition to oscillatory convection occurs. The additional parameters applying to the definition above are R, the zone radius; γ, the rate of change of surface tension with temperature; μ the dynamic viscosity; and κ, the thermal diffusivity. The experiment described above has motivated stability analyses of the half-zone flow fields which are described in the following sections.

Brief Introduction to Stability Theories

Stability theories have signification applications in fluid mechanics to determine flow states. The theories can be divided into two categories: energy theory and linear theory. Energy-stability theory provides a stability limit, under which a flow is guaranteed to be stable against any disturbances. The linear theory, on the other hand, gives a instability limit, above which, the flow is unstable, divided into two categories: energy theory and linear theory. Energy-stability theory provides a stability limit, under which a flow is guaranteed to be stable again regardless the magnitude of the disturbances. The linear theory, on the other hand, gives an instability limit, above which, the flow is unstable, regardless of the magnitude of the disturbances. Figure 2 shows a stability map with the two stability limits marked. The ideal situation, of course, would be for the two limits to coincide, but this occurs only in certain cases.

From the standpoint of the float-zone crystal-growth process, energy theory is an attractive technique, since it provides sufficient conditions for stability to disturbances of arbitrary amplitude. If the Marangoni number defined above was taken as the stability parameter, then energy theory provides e value Ma_E, such that for all $Ma < Ma_E$, stability is guaranteed. If the analysis was performed for an actual float-zone melt and if the crystal grower could operate the process to stay within the stability boundary, then striation-free material should be obtained. Energy theory is integral, or global in nature, defining stability in terms of the asymptotic decay of a disturbances-energy functional. Hence it provides good results in case where the mechanism responsible for the instability is of a similar global character.

Numerical Simulation of the Basic Flow Field

In order to perform energy stability analysis, the initial flow state must be determined first. To be related to available experiment results, a half-zone model of the float zones is adopted in this study. The flow domain in an actual half model experiment is very complicated. Figure 3 illustrates the simplified geometric and thermal conditions used in a half-zone model simulation. The basic state of interest is one of swirl-free, axisymmetric thermocapillary convection in a zone with aspect ratio of approximately 1. The momentum and energy equations of fluid flow have been solved numerically with certain assumptions, including nondeformable free-surface and flat interfaces between solid and liquid. The streamlines and isothermal (equal temperature) lines of a basic state in half-zone model are plotted in figure 4.

Energy-Stability Analysis

The energy-stability analysis begins with decomposing flow quantities, velocity, pressure and temperature, into two parts, basic state and disturbances:

$$Q = Q_{basic} + Q'$$

Substitution of the above expression into the governing Navier-Stokes equations and appropriate boundary conditions leads to a system of equations for the disturbance quantity Q'. The non linear disturbance-energy equation is derived from this set of equations. The basic state quantity Q_{basic} is coupled with Q' in this equation which has the following form:

$$\frac{dE}{dt} = -Pr * D + Ma * P + B$$

where the left-hand side is the change rate of the disturbance energy, Pr is a non-dimensional fluid property (Prandtl number); D is a "damping" term; and P is the "production" term; and B is the boundary-condition dependent term. All the terms are integrals of Q_{basic} and Q' over the flow domain. The dominant parameter is Ma. Ma_E is the value of Marangoni number under which the rate of energy change will always be negative, making the disturbances die away as time goes on. Therefore Ma_E is called the energy stability limit. As shown by the above equation, Ma_E depends on fluid parameter Pr and boundary conditions, among other factors. The equation has been solved numerically using a variational method, aided by an inverse-iteration technique for large sparse matrices. The solution was carried out on an IBM 3090 machine. Several assumptions were made in the solution process. One of them was that the disturbance to the flow was axisymmetric. This assumption allows the equations to be real, not complex, thus reducing the computer storage requirement. Another assumption was the nondeformable free surface, which reduced the number of equations to be solved as a coupled system. The results are compared with the experiment data and figure 5 shows the comparison. One notion here is that the experiment was performed using sodium nitrate fluid rather than silicon crystal. This is due to the fact that the molten silicon is an opaque medium and flow visualization is very difficult. The main difference between the two fluids, besides their visibility, is the Prandtl number. The Pr number of silicon is in the order of 0.01 and for sodium nitrate, it is roughly 7. The numerical results are obtained based as closely as possible on simulated experiment settings.

Conclusions and Discussion

There has been a large amount of analytic and numerical work on various aspects of thermocapillary convection. Most of the work provides examples of basic states. A Handful of stability analyses published were on simple flows with analytically defined basic state. The above energy-stability analysis is one of the first stability analysis using numerically determined basic states. Although it is limited by several assumptions because of the available computational resources, the results indicated two important things: (1) energy theory was capable of yielding sufficient conditions for stability of the right order of magnitude; and (2) the more difficult computations are needed to simulate the real flow.

Some follow-up extension work was done after the above study. One of these (ref. 3) has relaxed the assumptions about the disturbances, which improved the agreement with relevant experiment data. Another work in the field (ref. 4) included linear-stability analysis, which complements the results of energy-stability analysis. The combination of the linear limits and energy limit should provide an envelope of desired operating conditions under which the crystal should grow striation-free. However, the current agreement between the theoretic results and experimental data is not completely satisfactory because of the simplifications used in the numerical calculation. Possible steps for future research are: (1) take the influence of free-surface deformation into consideration; (2) modify the inadequacy in the half-zone model; and (3) model the full float-zone instead of the half-zone.

The results of stability analysis on a half-zone model of crystal growth process provided the insight on thermocapillary convection in melting crystals. Since the convection will be the dominant flow mode in microgravity environment, this type of study can be viewed as crucial for any space-based material processing to be successful.

Acknowledgments

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Biography

Yonghong Shen was born in China on Feb. 11, 1958. Shen completed her Undergraduate education, majored in Engineering, in 1982. After short period of working as teaching assistant at a university, she won a scholarship to continue graduate study at Stanford University. She obtained her Masters Degree in Mechanical Engineering in 1983. She then worked in various research and development institutes at Arizona and Illinois. In 1990, She came back to California to have a family. She joined Bentley Engineering Company in February of 1992, and worked on pressure system recertification (Code EEF, MS N213-8, Ext. 4-3035) since then. She is an associate member of American Society of Mechanical Engineers.

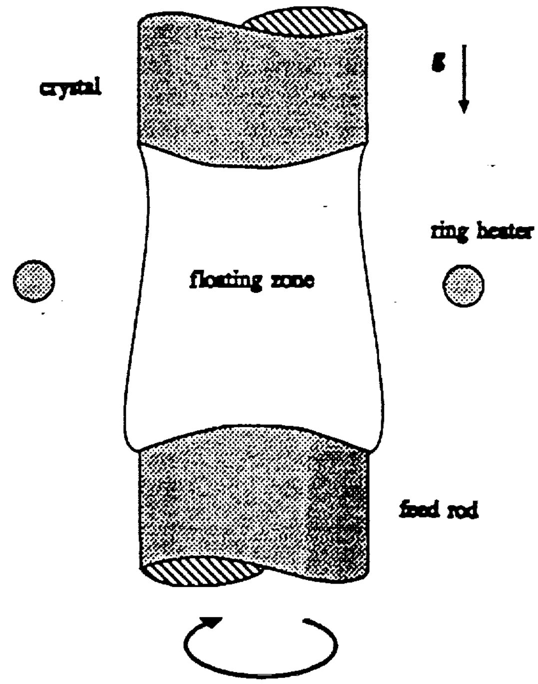

Figure 1. Schematic of the float-zone crystal-growth process

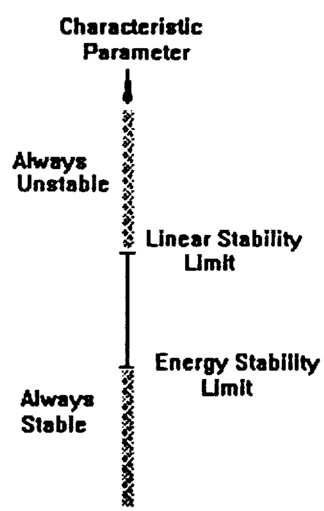

Figure 2. Stability map, showing the boundaries determined using energy and linear theories.

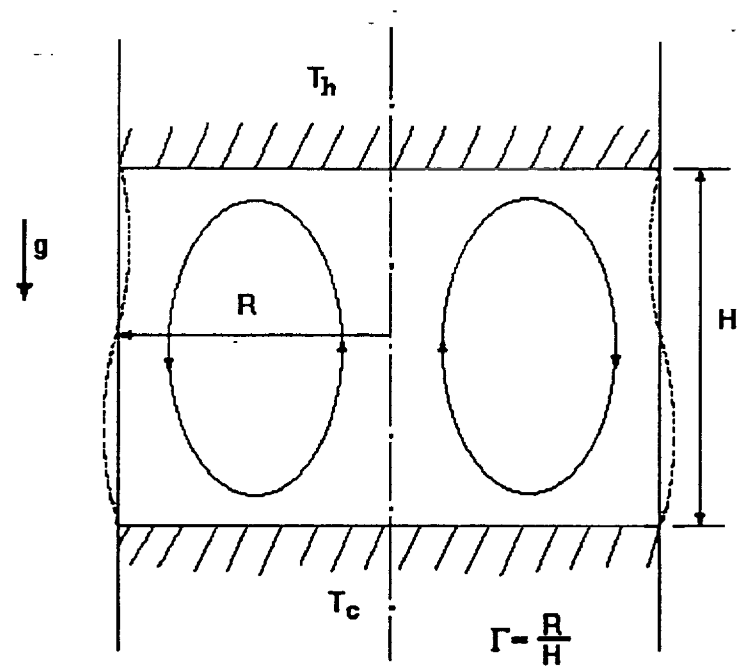

Figure 3. The half-zone model, showing geometric and thermal conditions.

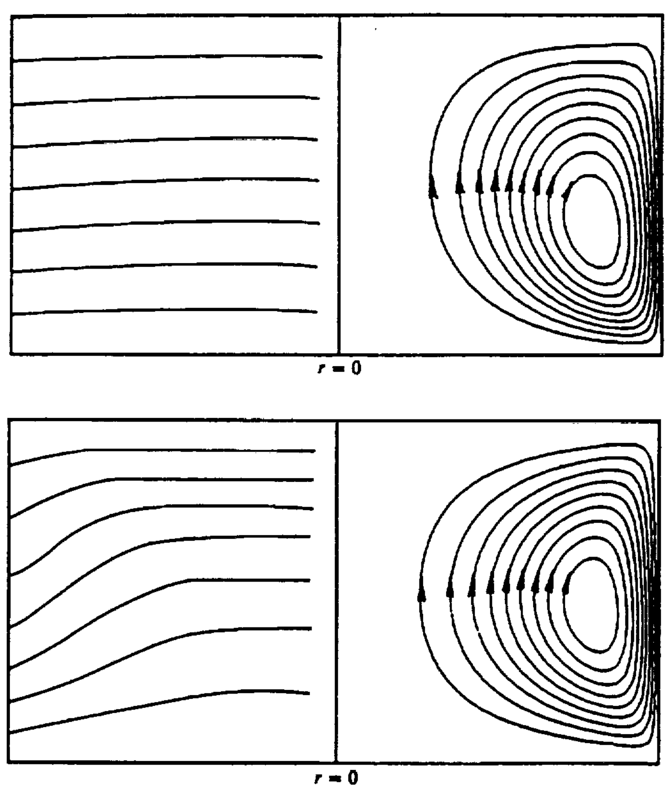

Figure 4. Sample basic-state isothermals and streamlines for aspect ratio G=1, Ma=100, and (a) Pr=0.01 and (b) Pr = 10.

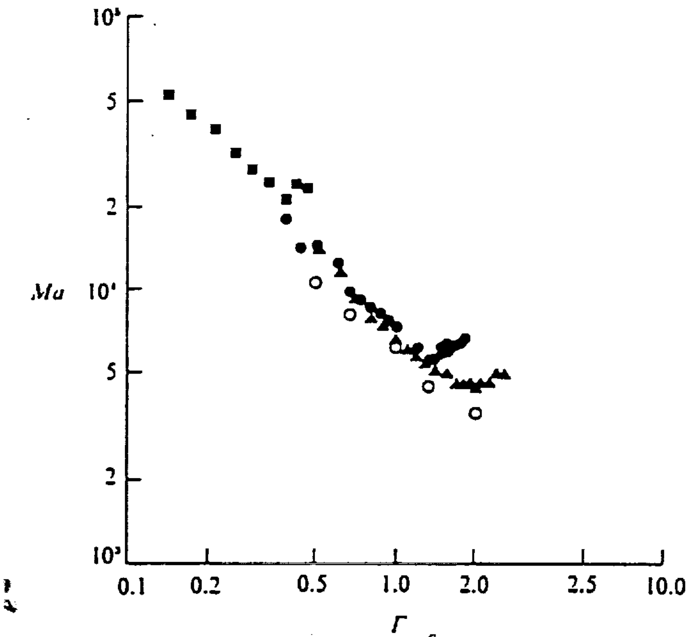

Figure 5. Comparison between result of present computation and model experiment (ref. 1) for various aspect ratios. Present: ⊙, Experiment: ▲ (radius=2mm), ● (3mm), ■ (10mm).