UTILIZING CONTROLLED VIBRATIONS IN A MICROGRAVITY ENVIRONMENT TO UNDERSTAND AND PROMOTE MICROSTRUCTURAL HOMOGENEITY DURING FLOATING-ZONE CRYSTAL GROWTH

Richard N. Grugel
Universities Space Research Association
c/o Marshall space Flight Center
MS - ES75
Huntsville, AL 35812

telephone: (256) 544-9165
fax: (256) 544-8762
e-mail: richard.grugel@msfc.nasa.gov

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ABSTRACT

It has been demonstrated in floating-zone configurations utilizing silicone oil and nitrate salts that mechanically induced vibration effectively minimizes detrimental, gravity independent, thermocapillary flow. The processing parameters leading to crystal improvement and aspects of the on-going modeling effort are discussed. Plans for applying the crystal growth technique to commercially relevant materials, e.g., silicon, as well as the value of processing in a microgravity environment are presented.

JUSTIFICATION FOR MICROGRAVITY RESEARCH

Gravity driven flow which occurs during float-zone processing is minimized in a microgravity environment and thus permits thermocapillary flow to be singularly investigated. Here, utilizing incremented and calibrated vibration, the consequence of flow velocities on microstructure can be controlled and systematically investigated, not just acknowledged. The microgravity environment will minimize unit-gravity induced biases such as static shape distortion and buoyancy flow. Here then is an opportunity to evaluate crystal growth and homogeneity in association with a stable and dimensionally optimized floating-zone.

BACKGROUND

The floating-zone technique for crystal growth of semiconductor materials has proven invaluable to the semiconductor industry. Unfortunately, significant convective flow in the liquid, both natural and induced is inherent to float-zone processing (Muhbauer *et al.*, 1983), both of which degrade crystal quality. Convection phenomena common to all floating-zone arrangements and relevant here are: A) surface tension induced flow (Marangoni or thermocapillary) and B) gravity induced flow (buoyancy). It should also be appreciated that flow induced by density differences can be minimized in a microgravity environment, whereas thermocapillary convection is *independent* of gravity level.

Many experimental and theoretical investigations have been conducted with the intent of understanding the influence of thermocapillary (TC) convection on crystal uniformity (e.g., Eyer *et al*, 1984; Eyer *et al.*, 1985; Cröll *et al.*, 1987). Schwabe *et al.* (1978, 1990) reported temperature oscillations in a $NaNO_3$ floating zone which was induced by surface flow and, based on a sounding-rocket experiment, also noted that oscillatory TC convection occurs in microgravity (Schwabe *et al.*, 1982). Jurisch and Loser (1990) and Jurisch (1990) showed striations of W in Mo single crystals which were attributed to oscillatory thermocapillary convection. A model (Young and Chait, 1990) where a floating-zone is simulated in a thin sheet in the absence of gravity revealed that convection and the presence of curved solid/liquid interfaces promoted lateral solute segregation; it was further suggested that a flat interface does not necessarily ensure uniform dopant distribution.

EXPERIMENTAL ASPECTS

Some studies toward understanding the effect of vibration on float-zone processing have been conducted (Anilkumar *et al.*, 1993; Grugel *et al.*, 1994; Shen *et al.*, 1996). A floating half-zone was simulated (Anilkumar *et al.*, 1993), Figure 1, by placing silicone oil between two vertical, 6.4mm diameter, aluminum rods which were separated by 2.5mm. Thermocapillary flow was initiated by imposing a temperature gradient (~20Kcm^{-1}) on the system via a resistance heater on the upper rod. As seen in Figure 1 top, this flow can be visualized by the addition of ~50μm diameter tracer particles which are illuminated by a He-Ne (10mW) laser sheet 0.5mm thick. In conjunction with the bottom rod is a vibrator that operates at a frequency of 70 Hertz and oscillation amplitude of ~100μm. The induced surface streaming flow, due to end-wall vibration of the float-zone, counteracts the thermocapillary flow which results in a balance, Figure 1 center. By increasing the vibration amplitude, the flow can be reversed (Figure 1 bottom).

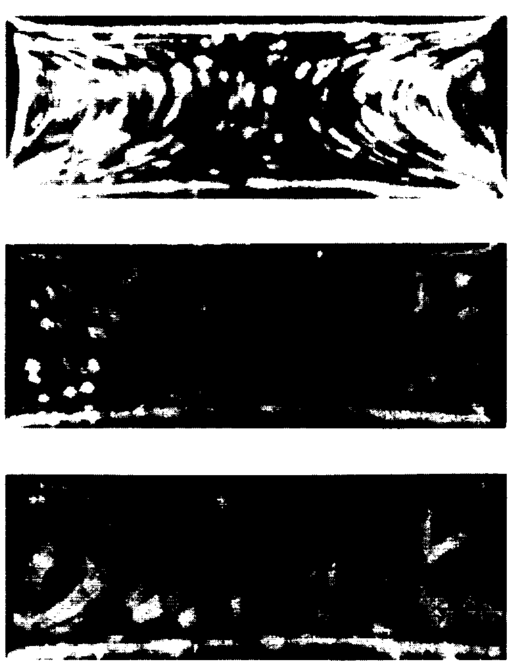

Fig. 1. Photographs (1/2 s exposures) of particle trajectories in the central section of a model half-zone of silicone oil (20cS) showing: (top) thermocapillary convection resulting from heating at the top, (center) balancing of the flow field through vibration of the bottom end-wall, and (bottom) reversal of the flow through imposition of higher vibration amplitude.

Using a fine (0.1mm dia.) k-type thermocouple it was also found that the radial temperature gradients smoothed considerably, thus suggesting a means to control thermocapillary flow and thereby improve crystal quality.

This innovative approach was expanded to investigate its effect in a traveling floating-zone of a model compound, sodium nitrate ($NaNO_3$) (Shen *et al.*, 1992). The choice of $NaNO_3$ has many advantages; when liquid allows direct observation and recording of flow patterns can be made, it has been successfully used in previous studies, and some of the thermophysical parameters are similar to a germanium or silicon melt. A traveling ring-heater assembly was constructed and used to float-zone process 6mm diameter $NaNO_3$ rods. When vibration, at a frequency of ~1.5kHz and amplitude of ~10μm, to the liquid zone was induced through the upper $NaNO_3$ rod the corresponding interface improved, i.e., became essentially planar, and flow decreased considerably. Again, radial temperature profiles improved and it was found that increasing the frequency effectively reversed the flow.

With these results, it was sought to demonstrate improved microstructural homogeneity. "Alloys" of the eutectic composition $NaNO_3$ - 18wt pct $Ba(NO_3)_2$ (Grugel *et al.*, 1994) were cast into 6mm diameter rods for float-zone processing with the intent of comparing distribution of the respective phases.

When the eutectic was solidified by the float-zone method, without vibration, at a rate $\approx$ 2.6mmhr^{-1} the $Ba(NO_3)_2$ phase is flake-like and distinctly finer in the center whereas on the sample periphery it is branched and coarser. When vibration is introduced to the system the solid/liquid interface, as in pure $NaNO_3$, flattens and subsequent examination reveals a uniformly coarse microstructure as a result of a uniform temperature gradient.

DISCUSSION

The effect of controlled vibrations on damping float-zone convection and improving microstructural homogeneity has been demonstrated. Still, considerable ground-based experimental and theoretical work is necessary. Here we will concentrate on defining the role various processing parameters play in promoting microstructural uniformity during floating-zone crystal growth. In particular float-zone dimensions, aspect ratios, oscillation frequency, and amplitude will be evaluated with the intent of understanding how thermocapillary flow is negated

In addition to experiments a comprehensive theoretical framework is being developed. An initial study has been completed (Lee, C.P., et al., 1996). This pointed out that the vibration-driven flow field is a subset of a large family of changeable flow fields set-up due to the non-linear oscillations of liquid bridges; flow fields changing direction and number of loops as the forcing frequency and/or the Reynolds number is changed. This streaming is due to the net body force imposed on the bulk of the column by the vibration.

Using the experimental parameters and properties of the silicone oil to calculate the Bond and thermal Marangoni numbers it was revealed that the latter, i.e., surface tension driven flow would clearly dominate over buoyancy. The impact of vibration on viscous flow can be estimated using the pulsating Reynolds number (Gershuni and Lyubimov, 1998). Finally the interaction between surface tension and imposed vibration can be characterized by the Weber number. These estimations of non-dimensional numbers are preliminary in nature; they are, however, very useful to ensure that the numerical model indeed represents the experimentally observed phenomena.

Transient numerical model was implemented using a finite element code (FIDAP, 1993). The typical one vortex structure of the flow with dominant thermocapillary convection (left side of the zone) is represented on Figure 2 and can be compared to Figure 1 (top). Dominant vibrational flow is shown on Figure 3. This flow has two vortices that represent wave motion initiated by moving rod. Small vortices close to the corner of computational domain on Figure 3 can be either secondary wave motion or flow generated by Schlichting mechanism (Gershuni and Lyubimov, 1998). Finally, Figure 4 pattern of reverse flow is presented, e.g., Figure 1 (bottom). It appears that this flow structure occurs within narrow parameter range.

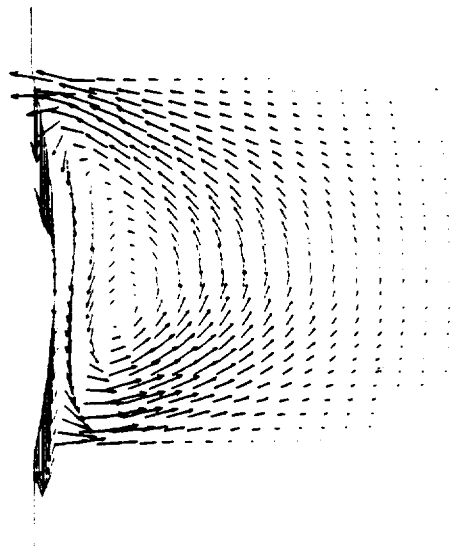

Fig. 3. Flow due solely to thermocapillary convection.

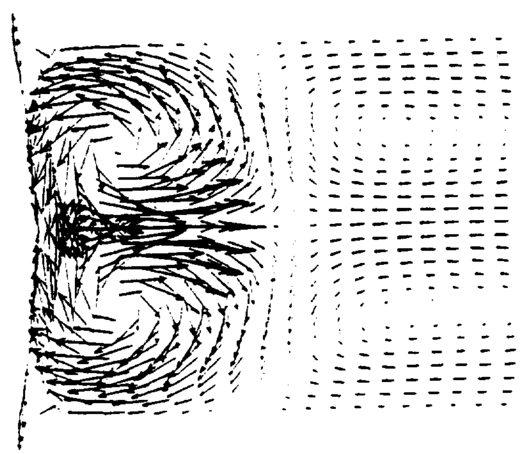

Fig. 4. Flow due solely to vibration at a moment when the rod movement is about to change direction.

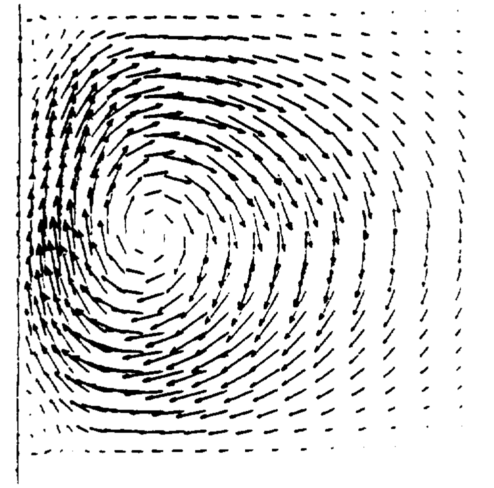

Fig. 5. Flow reversal due to interactions of thermocapillary convection and induced vibration.

Further investigation is necessary to accurately determine this range of parameters. The numerical data are in qualitative agreement with experimental findings; additional effort is required to obtain quantitative agreement.

Microgravity Extension

Gravity driven flow which occurs during float-zone processing is minimized in a microgravity environment and thus permits thermocapillary flow to be singularly investigated. Here, utilizing incremented and calibrated vibration, the consequence of flow velocities on microstructure can be controlled and systematically investigated, not just acknowledged. The microgravity environment will minimize unit-gravity induced biases such as static shape distortion and buoyancy flow; furthermore, sedimentation of tracer particles will be minimized. Here then is an opportunity to evaluate crystal growth and homogeneity in association with a stable and dimensionally optimized floating-zone.

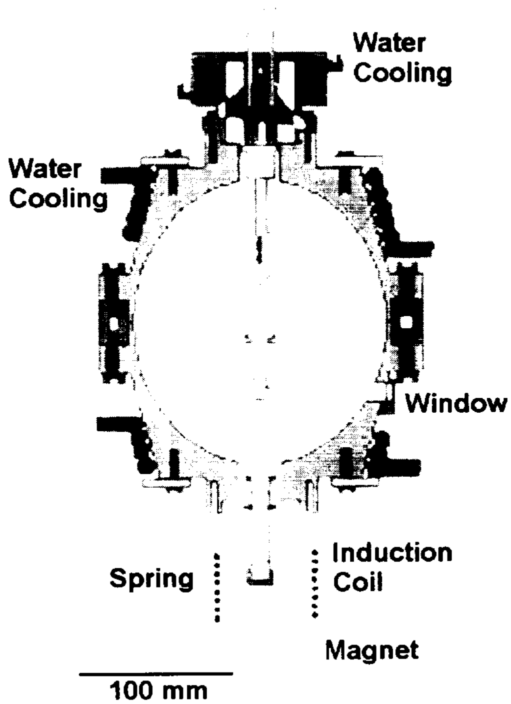

Fig. 5. Schematic of the monoellipsoid furnace (ELLI) with ampoule for float-zone experiments in conjunction with vibration.

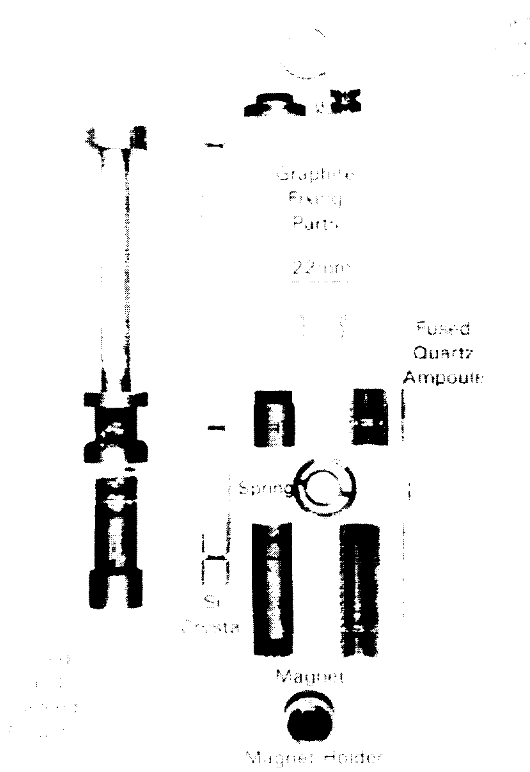

Fig. 6. Filled and sealed ampoule capable of inducing vibration to the liquid zone (left), and the individual components.

Investigating Si and Ge-based materials is an advantage in that thermocapillary convection dominates and promotes microsegregation even in a microgravity environment. Growth of the silicon and/or germanium-based semiconductors can be conducted in the Paraboloid-Ellipsoid Mirror Furnace (Elli) which was developed for growing crystals in space and used during the D2 mission, Figure 5. Figure 6 is a photograph of the components that will induce vibration to the growing crystal.

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