 FDMP, vol.1, no.1, pp.81-95, 2005

Thermocapillary Flow and Phase Change in Some Widespread Materials Processes

Gustav Amberg[1], **Junichiro Shiomi**[2]

Abstract: A few issues in materials science are reviewed with regard to the importance of fluid flows. The effect of convection on generic solidification problems is discussed. One relevant class of flows in melts is those driven by surface tension gradients. In welding this thermo- or solutocapillary flow will determine the penetration depth, and will depend very sensitively on the composition of the material, through the dependence of surface tension on temperature, presence of surfactants, etc. In crystal growth the convective motion in the melt may cause instabilities that are often undesired in practical processes. The unsteady flow structure can cause inhomogeneous chemical composition at the solidification interface. Work has recently been done to apply active feedback control to suppress the thermocapillary oscillation. In the high Prandtl limit, a significant attenuation can be obtained by means of rather simple control methods.

keyword: Thermocapillary, convection, float zone, control, welding, dendrite

1 Thermocapillary Convection

Thermocapillary convection becomes important whenever a strong temperature gradient is present over a free liquid surface, in particular if the surface tension dependence on the temperature is strong. Also, if the dimensions of the liquid volume are small, surface forces will be relatively more important compared to volume forces. These conditions are frequently satisfied in materials processes such as welding and crystal growth. Also in microgravity experiments with fluids, thermocapillarity may typically be the dominant source of convection.

Some light can be shed on the intrinsic nature of thermocapillary convection by considering a small section of a free liquid surface with a temperature dependent surface tension,

$$\sigma = \sigma_0 + \gamma \cdot (T - T_0), \tag{1}$$

in the presence of a temperature gradient along the surface. In the typical situation of a surface tension that decreases with temperature ($\gamma < 0$), the surface tension thus increases in the direction of decreasing temperature along the surface. Considering a force balance over a thin control volume containing the free surface it is clear that the difference in surface tension must be balanced by a shear stress in the fluid. The typical scenario picture is thus that the fluid surface is dragged towards cold spots at the surface. Thermocapillary convection is often characterized by the value of the Marangoni number, defined as $Ma = \gamma \Delta T L/(\alpha \mu)$, i.e. a Peclet number based on the thermocapillary velocity scale $\gamma \Delta T/\mu$. Here, ΔT is the characteristic temperature difference, L is a length, α thermal diffusivity, μ the dynamic viscosity.

The above would typically be true for pure fluids. In the presence of a possibly surface active additive, concentration gradients would drive a *soluto* capillary flow in a similar way. However in the presence of a surfactant, which is concentrated on the surface, there is a more complicated coupling between the flow field and the surface tension: Consider a stagnation point flow on the surface, where fluid rises to (descends from) the surface and spreads out (converges) along the surface. The surface is thus stretched (contracted) and the local surface concentration of surfactant would tend to decrease (increase). As the concentration decreases (increases), the surface tension increases (decreases), and thus a restoring force appears. This effect could be termed surface 'elasticity'. Also, depending on the properties of the surfactant, the surface may have dilational and shear viscosity to various degrees.

[1] KTH Mechanics, S-100 44 Stockholm, Sweden, gustava@mech.kth.se
[2] Department of Mechanical Engineering, The University of Tokyo 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-8656, Japan

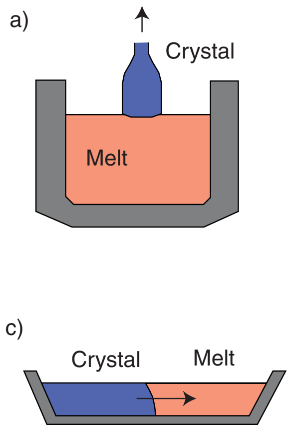

Figure 1 : Sketches of the geometrical configurations in a) Czochralski growth, b) floating zone, c) Horizontal Bridgeman growth.

1.1 Model problems inspired by crystal growth

When growing crystals for electronic and optical purposes, the challenge is frequently to produce a single crystal of precisely controlled and highly uniform properties. This is presently a subject of obvious and growing importance for electronics, optics, laser technology and other applications, with the largest area being semicondutors accounting for 60% of the 20000 tons crystals produced in 1999, Scheel (2000).

There are many different processes that are used, see Scheel (2000) for a recent review, but here we are concerned with methods where the crystal grows by solidification from a melt, where convection may have desirable or undesirable effects, Langlois (1985). Sketches of a few common geometric arrangements are shown in fig 1. The most important method, economically, would be Czochralski growth where the heated melt is kept in a crucible, and a crystal is pulled up by slowly raising a cooled seed crystal in contact with the melt. In horizontal Bridgman growth, the melt is contained in a boat, which is moved in a temperature gradient, so that the melt solidifies in a controlled manner. In the float-zone method, an intense heat source is passed along a polycrystalline rod, so that the material melts and re-solidifies as the heat source passes. There is thus a liquid bridge of melt, which is held by surface tension forces between the two solid ends of the rod. This method has the advantage that it is containerless and thus holds a promise for very pure crytals, but it is complicated by the fact that gravity may have a large influence on the suspended melt drop.

A common feature of these methods is the presence of a free surface subject to relatively high temperature gradients. Hence, this is indicative of the possible importance of thermocapillary convection. Also, the heat and mass transfer in the melt determines the homogeneity of the finished crystal. The importance of thermocapillary convection in such systems was pointed out already by Chang and Wilcox (1976), Chun and Wuest (1979), Schwabe and Scharmann (1979). Numerous studies have since then investigated different aspects of thermocapillary convection in configurations resembling crystal growth processes. A broad overview of this general area is given in Lappa (2004a, 2005b). The studies reviewed below deal with axisymmetric geometries. Related phenomena in cubical and parallelepipedic containers are reviewed by Lappa (2005a), in this issue.

1.1.1 The Floating-Zone

A common model problem is the half-zone, see figure 2b. In this configuration, a liquid drop is held between two circular cylinders by surface tension forces. The flow is driven by a temperature difference between the two rods causing a temperature gradient, and hence a surface tension gradient, along the free surface. Such a model is then thought to represent half of a real zone, which is heated at the center, giving rise to two recirculating flows, one above and one below the heat source. The main problem with the Half Zone model is that it disregards possible interaction between these two zones. Lappa (2003, 2004b) clearly illustrated that there are qualitative and quantitative differences between the half and the real floating zone of full extent.

However, much work has been invested in understanding the half zone case, in view of its relative simplicity, and possibly also because the design of well defined thermal boundary conditions in experiments is more straightforward. The main objective of most studies related to the half-zone has been to understand the stability characteristics of the steady basic thermocapillary flow. The motivation for this is the observation that crystals grown with the floating-zone method typically have periodic axial variations in dopant concentration, so called striations, which are attributed to oscillatory thermocapillary convection in the melt. The relevant fluid mechanical problem is thus the understanding of the flow instabilities that lead to the aforementioned unsteady motion.

The fundamental instability mechanisms in thermocapil-

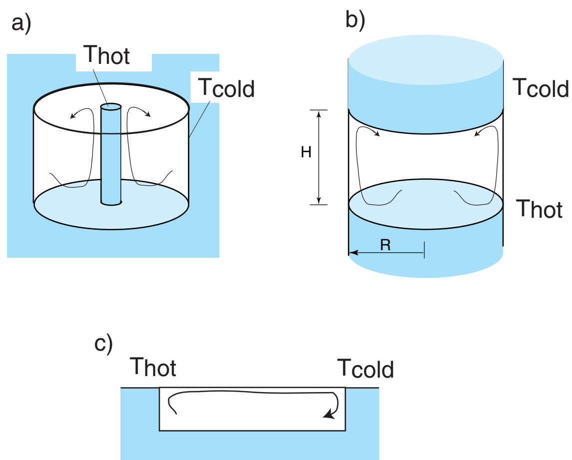

Figure 2 : Sketches of a few model problems related to crystal growth a) Annular configuration , b) Half zone, c) Horizontal rectangular cavity.

lary flows in even simpler geometries, i.e. infinitely long plane layers and cylinders, were studied by Smith and Davis (1983), Xu and Davis (1984) and Smith (1986), who identified among other things the fundamental thermocapillary wave instability. Theoretical studies of stability in half zones include studies of linear and energy stability theory by Shen, *et al.* (1990), Neitzel, *et al.* (1991); Neitzel, *et al.* (1993); Wanschura, *et al.*(1995); Chen, *et al.* (1997); and Levenstam, *et al.*(2001). Full numerical simulations of the developed instability have been made by, among others, Rupp, *et al.* (1989); Levenstam and Amberg (1995), and Levenstam, *et al.*(2001).

Experimentally, the half-zone has been a popular model geometry, investigated first by Preisser (1983). See also the recent review by Schatz and Neitzel (2001). It is quite difficult to carry out well-controlled experiments using technically interesting fluids with small Prandtl numbers such as semiconductor melts. Instead, a large literature has appeared where thermocapillary flows were studied using fluids with Prandtl numbers greater than one, typically silicone oils or molten salts. Early such studies are those by Preisser, *et al.*(1983) and Velten, *et al.*(1991). There have been attempts to measure the stability characteristics of flow in systems resembling real float-zones, using real semiconductor materials, Cröll (1989, 1991) and Levenstam, Amberg, *et al.*(1996), but when trying to understand the fundamental instabilities, these experiments are hampered by uncertainties in the material properties, difficulties in visualizing the flows, etc.

Another geometry where the dynamics have similarities with the half zone is the annular geometry shown in figure 2a. In this case, a cylindrical container with a small co-axial cylindrical heater is used. The fluid is contained in the annular gap between the heater and the container wall. A free surface subjected to a radial gradient of temperature is hence created. Kamotani, *et al.*(1992) were the first to experimentally study a thermocapillary flow in a cylindrical container of the annular type. This geometry is attractive since it presents many experimental advantages, and the dynamics can be expected to be similar to other axisymmetric thermocapillary convection cases. More recent microgravity experiments by Kamotani (1997,1998,2000) investigated the onset of oscillations in this geometry using silicone oil with Prandtl number around 27.

The picture that emerges is that, unfortunately but not very surprisingly, the quantitative and qualitative features of the oscillatory flow depend strongly on the Prandtl number of the fluid. Figure 3 shows the critical thermocapillary Reynolds number $Re = Ma/Pr = \gamma(T_{hot} - T_{cold})H/(\nu\mu)$ vs Prandtl number $Pr = \nu/\alpha$ for the half zone problem in fig 2b (Levenstam, Amberg, and Winkler, 2001). In low Prandtl number fluids below 0.05, corresponding roughly to interesting metal and semiconductor melts, the flow becomes oscillatory at a $Re \approx 6000$, independent of Pr. In this Prandtl number range, the onset of oscillations is thus an entirely inertial hydrodynamic instability, Levenstam and Amberg (1995), Wanschura, *et al.*(1995), and the proper parameter for characterizing the instabilities at low Prandtl numbers is thus the thermocapillary Reynolds number (defined as $R_l = Ma/P_l$), rather than the Marangoni number. In the high Prandtl number range, above Prandtl numbers about 1, the mechanism is quite different and involves a complicated coupling between the temperature and velocity disturbances related to the thermocapillary wave, Wanschura, *et al.*(1995). The critical Reynolds number continues to decrease with increasing Prandtl number, while a corresponding critical Marangoni number ($Ma = Re \cdot Pr$) increases.

It is interesting to note that in the region with Prandtl number just below unity (actually $0.05 < Pr < 0.8$), the axisymmetric flow is much more stable than outside of this range, with critical Reynolds numbers around ten times larger than the levels outside. This is due to the

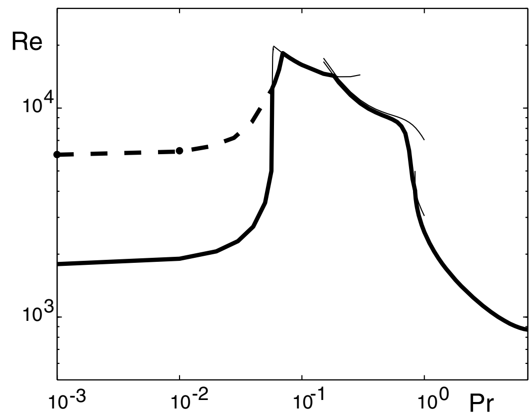

Figure 3 : Stability of the flow in a half zone. Below the lower curve the flow is steady and axisymmetric. At Prandtl numbers above 1, the flow is oscillatory above the curve. At low Prandtl numbers the flow is steady and 3D between the full and the dashed line, and oscillatory above the dashed line.

fact that the action of the thermocapillary stress changes qualitatively with the Prandtl number here; In the low range, when convection of heat starts to be important at $Pr \approx 0.06$, the thermocapillary stress generated by the inertial instability present there, is actually a restoring force that counteracts the instability. This competition stabilizes the flow and raises the critical Reynolds number by an order of magnitude, and gives rise to a complicated sequence of critical modes as the Prandtl number is increased. When the inertial instability loses its importance to a thermocapillary instability mechanism around $Pr \approx 0.8$ the critical Reynolds number again drops dramatically. Similarly it can be seen that the thermocapillary stress related to the disturbance is now destabilizing.

In this context, it is natural to try to apply active feedback control to suppress oscillations. Here the idea is to attenuate the oscillation by altering the thermocapillary instability without influencing the base flow appreciably. The input to the control law is usually a measurement of the local surface temperature, and the output is heat flux added (subtracted) by heaters (coolers) at the surface. Since the surface temperature distribution plays a key role in the instability mechanism, it should be efficient to influence the oscillation via the surface temperature.

There is another type of control method that delays the onset by reducing or altering the basic flow state. It decreases the effective Marangoni number. For example, a well known method is to apply a magnetic field to an electronically conductive melt (Leon *et al.* 1981, Kimura *et al.*, 1983, Robertson and O'Conner, 1986a,1986b). Others methods counteract the surface flow by generating a steady-streaming type of flow by applying an end-wall vibration (Anilkumar *et al.*, 1993, Shen *et al.*, 1996, Lee *et al.*, 1996, 1998), or directing a gas jet parallel to the surface (Dressler and Sivakumaran, 1988). A drawback of these methods is that the damping of the base convection enhances the macro-segregation of the chemical compositions due to the weakening of the global mixing. Compared with these base flow control methods, the active feedback control may be beneficial in terms of both microscopic and macroscopic homogeneity of the final single crystal. However, there has been only a limited number of studies reported in the literature focused on such a strategy.

In half-zone model experiments, Petrov *et al.* (1996, 1998) attempted to stabilize the oscillation by applying a nonlinear control algorithm using local temperature measurements close to the free surface and modifying the temperature at different local locations with Peltier devices. They constructed a look-up table based on the response of the system to a sequence of random perturbations. A linear control law using appropriate data sets from the look-up table was computed. The control law was updated at every time step to adapt the control law to the nonlinear system. Using one sensor/actuator pair, successful control was observed at the sensor location for $Ma \sim 17750$. However, infrared visualization revealed the presence of standing waves with nodes at the feedback element and the sensor. This was resolved by adding a second sensor/actuator pair, enabling the control to damp out both waves propagating clockwise and counterclockwise, and thus standing waves. The performance of the control was reported for only one value of $Ma \sim 15000$, where the critical value was $Ma_{cr} \sim 14000$. They stated that the oscillation could not be suppressed when Ma exceeds the critical value by more than 8.5%, mostly due to the weak response of the fluid flow to the Peltier devices, which cannot be cooled more than a few degrees during the application of the control pulse.

A simpler and perhaps more robust proportional feedback control method was demonstrated by Shiomi *et al.*(2001). The feedback control was realized by measuring temperatures at different locations, and using these to modify the surface temperature via a simple control law. It is worthwhile to stress how Strategically placing sensor/actuator pairs (controllers) and using the knowledge of the modal structures, a simple cancellation scheme can be constructed with only a few controllers. A series of articles explore the possibility of applying the control in simplified geometries such as the annular configuration and the half-zone for high Prandtl number liquids by means of experiments (Shiomi *et al.*, 2001, 2002, 2003, Bárcena *et al.*, 2005), numerical simulations (Shiomi and Amberg, 2005a), and formulation of a simple model equation system (Shiomi and Amberg, 2005b).

Successful suppression of the oscillation was obtained especially in the weakly nonlinear regime where the control completely suppresses the oscillations. Typical pictures of the successful control of a weakly nonlinear thermocapillary oscillation are shown in figure 4. The top (bottom) figure depicts the time history of a temperature (power output) signal at a sensor (heater) location. With a proper choice of actuators, even with the local control, it was shown that it is possible to modify the linear and weakly-nonlinear properties of the three-dimensional flow system with linear and weakly nonlinear control. On the other hand, the method exhibits certain limitations. Depending on the geometry of the system and actuators, the limitation can be caused by either the enhancement of nonlinear dynamics due to the finite size of the actuators or the destabilization of new linear modes. The former case can be attenuated by increasing the azimuthal length of the actuators to reduce the bandwidth of the wavelengths of waves that are generated. In the latter case, having an idea of the structure of the new destabilized modes, they can be delayed by optimizing the configuration of controllers. On the whole, the oscillation can be attenuated significantly in a range of supercritical Ma up to almost twice the critical value.

Practical floating-zone processes and experiments are (naturally) more complicated than the half zone and annulus discussed above. The temperature differences that are used are such as to give thermocapillary Reynolds numbers of the order $O(10^5)$ rather than the $O(10^3) - O(10^4)$ discussed so far. Experimentally a 'periodic' and a 'turbulent' regime has been observed, Cröll *et al.* (1991), in terms of the regularity of the observed striation patterns in the finished crystals. Lan *et al.* (2000) have recently presented a simulation of a full Si float zone at a realistic Reynolds number of 10^5, showing a growth

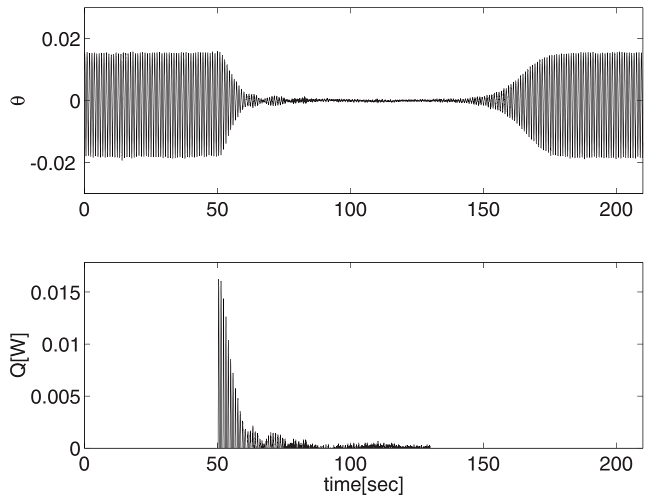

Figure 4 : A typical picture of successful control of thermocapillary oscillation with weak nonlinearity in a half-zone experiment. Top: Time history of the dimensionless temperature signal. Bottom: Simultaneously measured heater output power. Marangoni number is 18 percent above the critical value.

speed that is apparently chaotic in time. Model experiments that investigate the chaotic regime in a systematic fashion have appeared recently, Ueno *et al.* (2000), Kawamura *et al.* (2001), who carried out experiments with silicone oil in a half-zone with a driving temperature difference up to 100 K, i.e. 4-5 times the critical value.

The flow in a real float zone is highly chaotic, even if the Reynolds number is hardly high enough for engineering turbulence models to give good results. In practice there are also other possible sources for unsteady flow, such as RF-inductive heating, and buoyancy driven convection (when present), even if the basic thermocapillary mechanisms discussed above are generally regarded as the most severe. In order to stabilize the flow, differential rotation of the rods, magnetic fields etc, may be applied (see, e.g., Lan and Yeh 2005). Quantitative simulations must also account for the deformation of the free surface, the evolution of freezing and melting interfaces, etc. In addition to those referenced above, Rao and Shyy (1997), Kaiser and Benz (1998), Ratnieks *et al.* (2000) may be mentioned.

1.2 Czochralski growth

In the Czochralski system, see figure 1a, a melt is kept in a crucible that is typically inductively heated. The crystal is grown by cooling it and slowly pulling it upwards, so that new material solidifies. This process is widely used in industry for the production of single-crystalline silicon for electronics, where the sizes of the crystals have grown dramatically in recent years, to 8" diameter presently. Some of the issues in this process are the uniformity of the crystal, notably oxygen content, and the presence of crystal defects that may be related to thermal stresses in the crystal. As in the float zone, the heat and mass transfer in the melt is crucial in this regard. In addition to natural and thermocapillary convection, the crucible (container) and the crystal are usually rotated as a means of influencing the mean flows, which is another important source for melt motion.

Much work has gone into setting up complete models of this important industrial process. In order to be quantitative these need to include the entire furnace to capture the radiation heat transfer at high temperatures, as well as the influence of the gas motion above the melt, etc, see Dornberger *et al.* (1997), Zhou *et al.* (1997), and Chatterjee *et al.* (2000), Tsukada *et al.* (2005). There are thus many other complications, in addition to the melt flow, but still this is often identified as the main remaining obstacle. The Czochralski systems are considerably larger than the floating zones discussed above and the melt flow is typically closer to proper turbulence. Orders of magnitudes of parameters relevant for the flow, Lipchin and Brown (1999), could be Grashof numbers $\approx 10^{11}$, and Prandtl number 0.011, indicating that buoyancy alone would be strong enough to cause a turbulent flow.

Classical turbulence models have been employed in this context, see for instance Lipchkin and Brown (1999), who compare systematically different wall treatments for the k-ε model. According to their analysis, the melt flow in Czochralski growth has many of the features that are notoriously difficult to treat using k-ε models; Rather low Reynolds number flows, natural convection, separation, effects of system rotation, etc. Large eddy approaches have also been tried, Basu *et al.* (2000), Evstratov *et al.* (2000), even though the resolution is typically not very high, in view of all the additional complications in this problem.

Czochralski processes are also used for growing crystals from oxide melts, as for instance YAG ($Y_3Al_5O_{12}$) for laser applications. Such melts typically have a Prandtl number around 10, and are more viscous than say a silicon melt. The resulting flows may have typical values of Grashof numbers of the order $10^4 - 10^7$, Marangoni numbers in the order $10^3 - 10^5$, etc, and are thus not necessarily turbulent, but may show oscillatory motions and low dimensional chaos, Jing (2000), Enger *et al.* (2000), Xiao and Derby (1994), Tsukada *et al.*(2005).

2 Welding

Gas-Tungsten-arc (GTA) welding is a widely used method to join materials in manufacturing industries. Nevertheless, the physical processes involved in GTA-welding are highly complex and are not fully understood. One key issue in improving welding technology is to devise methods suitable for new materials, and to predict the welding properties of a new material in detail. This involves for instance a prediction of the depth and width of the molten region (the weld pool), the structure of the material in the junction after the process is complete, and also how the properties of the material influence the choice of the actual welding parameters, i.e. welding current, speed, etc. There has recently been a growing interest in detailed numerical simulations of weld processes (see the recent conferences, Cerjak (2001), and David and Vitek (1993)).

A generic case for studying weld pool phenomena would consider a flat plate and an electric arc struck between an electrode above the surface and the plate. The molten weld pool develops directly beneath the electrode when the current is turned on and its shape and size are highly influenced by the heat and fluid flow in the molten zone.

The fluid flow in the weld pool is mainly driven by forces due to surface tension gradients (the Marangoni convection), but is also strongly influenced by electromagnetic forces and buoyancy, Mundra and DebRoy (1993a,1993b), Oreper and Szekely (1984). Arc pressure and aerodynamical drag forces arising from the shielding gas used in GTA welding to prevent oxidation have an impact on the welding process. Moreover, heat losses due to radiation and convection and solidification of the weld fusion zone as well as the modeling of the heat input from the arc present between electrode and workpieces have to be taken into account. The process is also highly influenced by the presence of surface active elements on the surface of the melt. In the case of stainless steel, sulfur and oxygen are known to be surface active.

Figure 5 shows stationary temperature and velocity distributions for a 3D GTA-welding simulation of a repre-

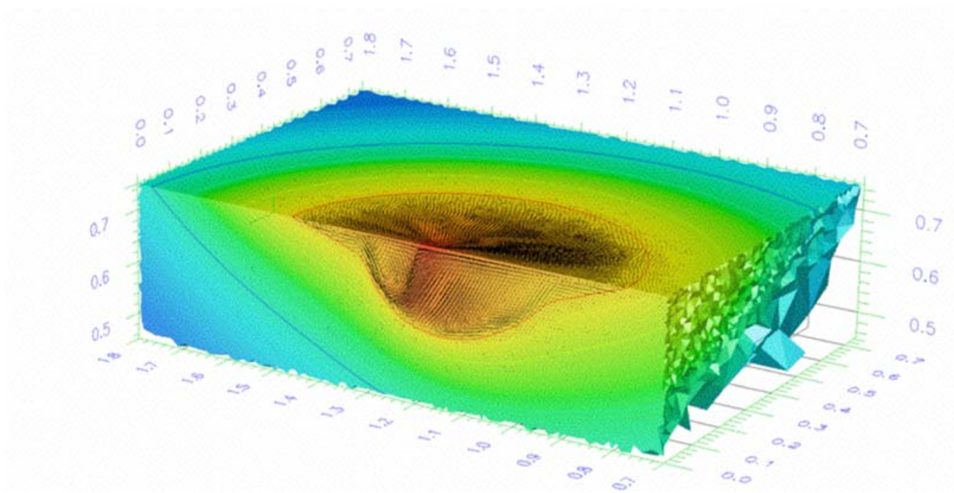

Figure 5 : Isotherms and flow vectors on the surface and symmetry plane of a plate during welding.

sentative welding situation, Do-Quang (2004). The electrode is moving with a constant speed of $6mm/s$ in the positive y-direction, to the left in the figure. The heat flow from the electrode to the workpiece melts a specific region of the specimen which forms what is called the weld pool. The shading indicates the temperature distribution on the upper surface of the plate and on the vertical symmetry plane oriented along the traveling direction of the electrode. In these planes also the velocity fields are plotted. The melt-solid interface is indicated as a contour separating the solid no-motion areas from the melt region. It is observed that the flow pattern has a fore-aft asymmetry, due to the motion of the electrode. The temperature fields are strongly affected by convection, with characteristic velocities of $0.1m/s$. The fluid flow in the weld pool is highly complex, and the heat transfer that results determines the weld pool depth and shape. Moreover, the velocity field at the surface of the specimen determines the streamlines defining the traveling paths of, for example, slag particles. In this particular case a steel with a comparatively large sulfur content is modeled, for which surface tension increases with temperature. This results in melt at the surface being pulled by surface tension gradients inwards to the center. This gives rise to a beneficial flow pattern, with a jet directed downwards from the hottest spot at the center. This hot jet will cause the material to melt, and form a deep and narrow weld pool.

In order to study quantitatively the mathematical modeling of phenomena in a weld pool, Winkler et. al. (1998, 2000a,2000b) made well controlled experiments on a

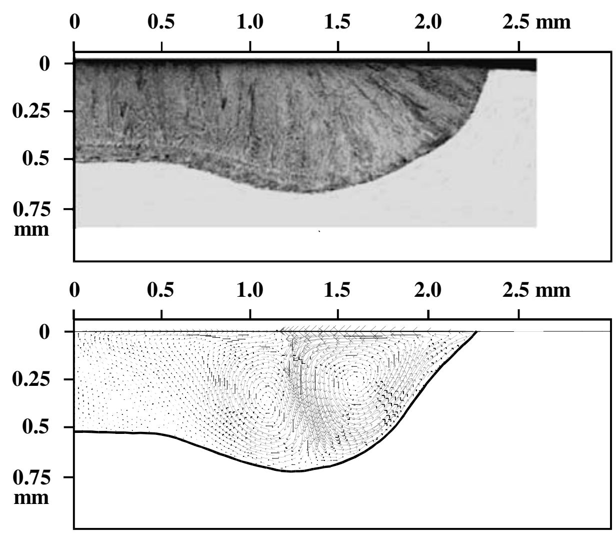

Figure 6 : Crossection of an axisymmetric point weld, comparison between the experimentally obtained structure, revealing the shape of the largest liquid region, and the simulated weld pool shape and velocity field. The horizontal axis is radial distance from the axis of symmetry, and the vertical is distance from the upper surface of the plate.

spot weld, i.e. using a stationary electrode and heating a stainless steel plate of well controlled composition, during a specific time, typically a few seconds. When the arc is removed the weld pool quickly resolidifies and its final shape can be observed from the microstructure of the material when the plate is cut through the weld. The weld pool shapes obtained thus were compared to timedependent axisymmetric numerical simulations using a comprehensive mathematical model. Figure 6 shows such a comparison for a stainless steel with low sulfur and oxygen content, that has been subjected to a stationary heat source for 1s. It should be noted that the agreement is quite satisfactory, both quantitatively and qualitatively. The weld has in this case developed into a rather complex shape, with the maximum depth appearing at about 1.2 mm from the center, despite the fact that the heating intensity is maximal at the centerline. This is characteristic of materials with very low presence of surface active elements such as sulfur and oxygen. In such systems, the surface tension is decreasing with temperature over most of the temperature range, causing the largest surface tension to appear near the rim of the pool. This gives rise to a rather unexpected flow field, with a vortex pair transporting hot melt from the surface down to the solid at this distance from the center. This peculiar flow pattern is the result of the competition between the different driving forces for convection, i.e., surface tension gradients, electromagnetic force, buoyancy, etc. The surface tension depends on temperature as well as composition, and it can be seen that the surface tension has a local maximum at the point where the flow turns downwards from the surface.

The detailed modeling of surface tension is thus crucial for accurate results. In the past, equilibrium models have been used that assume that the surfactant concentration at the surface is in perfect equilibrium. When using such models to evaluate tests like that in figure 6 it was found that the nominal surfactant concentration had to be adjusted in order to make the model fit. This was interpreted as an indication of a redistribution of surfactants by the convective flow. Thus, using a nonequilibrium model that accounts for the surface elasticity described before, together with a crude calculation of the mass transfer of the surfactants in the weld pool, good agreement was obtained for a variety of experimental conditions.

Figure 7 shows the computed surface concentration of sulfur together with the flow field, as viewed from above, for a case with a moving electrode (Us = 20cm/min, $I = 200$A, $U = 10.2$V). The motion of the electrode is upwards in the figure, causing a characteristic drop-shape of the weld pool, with a blunt leading edge, and a pointed trailing edge. The heat source is located at $x = y = 0$, in the upper part of the figures. The steel in this case also has a low level of surfactant concetration, so that over most of the surface the surface tension is decreasing with temperature. The corresponding flow pattern on the free surface is directed outward, away from the heat source. In the rear part of the pool however, there is a distinct stagnation line separating a trailing region where flow velocities are smaller, and actually are reversed. The stagnation line in the velocity clearly coincides with a band of increased sulfur concentration, as shown in the left panel. As expected, sulfur is accumulated at a converging stagnation line. Conversly the sulfur concentration is decreased over the central part where the surface flow is diverging.

3 Solidification

One crucial step in almost all materials processes is solidification in one form or other. The conditions under which the melt resolidifies will be crucial for the final microstructure of the material. The size and morphology of the individual grains that make up a polycrystalline material, the homogeneity of a monocrystal, the actual phase that is formed, as well as its local composition, are determined by the interplay between local heat and mass transfer and the thermodynamics of the phase change. Even though the microstructure of the material may change considerably during subsequent cooling and following process steps, the foundation has been laid at the point of solidification. Since local heat and mass transfer governs the phase change, it is obvious that any melt convection at all will be of paramount importance in determining the structure of the material, thus making this an area of important applications that should interest fluid mechanists.

3.1 Stability of a solidification front, dendrites

A generic example of solidification of a pure liquid would be the unidirectional solidification of an undercooled sample initially at a temperature below the freezing point. The simplest mathematical description of this would assume a planar phase change boundary, with a

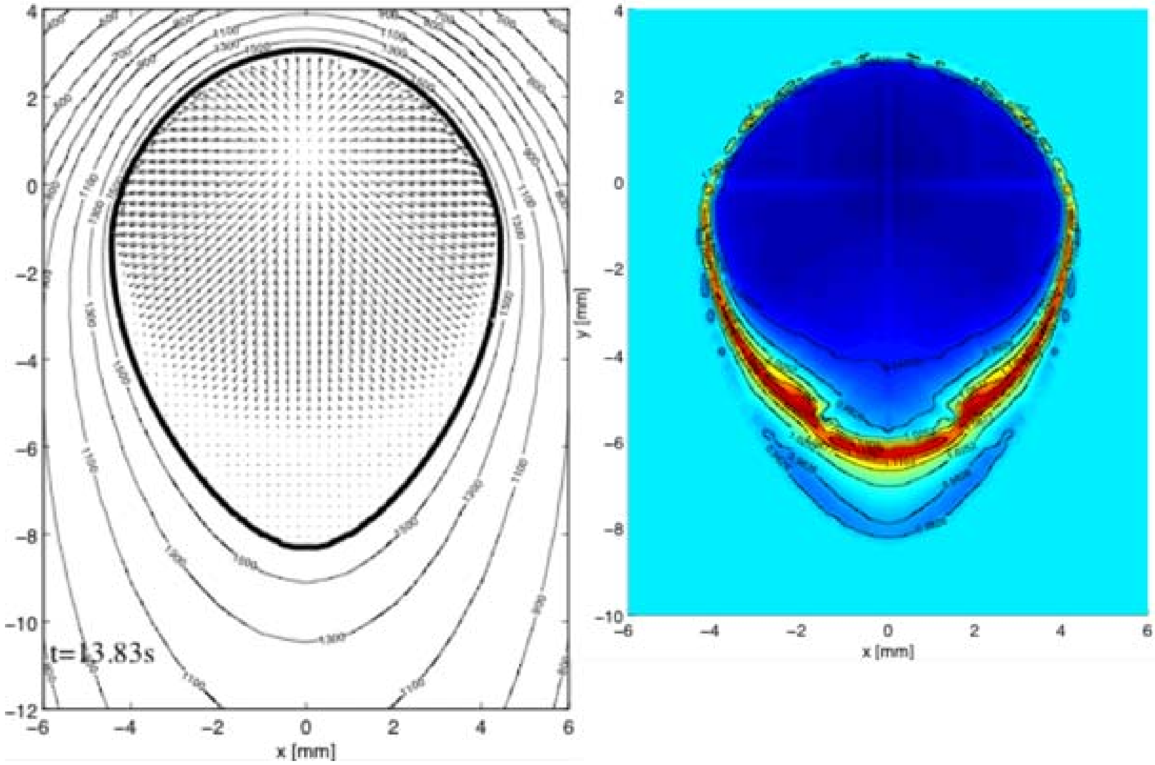

Figure 7 : Profiles of the computed surface concentration of surfactants.

constant given freezing temperature at the solidification interface, and a constant latent heat release expressed as a discontinuity of the normal temperature gradient at the interface. This evokes a picture of the solidification front as a smooth interface advancing over the domain. This however is very much the exception rather than the rule when dealing with metals and crystalline materials.

The reason for this is that a planar solidification interface advancing into an undercooled melt is subject to a fingering instability very similar to fingering in Hele-Shaw cells: if a bump is formed on the solidification front, the local temperature gradient ahead of it will increase, and thus cool the front more efficiently there, causing an amplification of the disturbance. Furthermore, this mathematical problem is ill-posed, since the growth rate of a disturbance of the planar shape of the interface will grow unboundedly with the wavenumber of the disturbance. The assumption responsible for this is that the temperature is assumed constant on the interface - in the Hele Shaw analogy this would correspond to a zero surface tension. A more realistic description of solidification is obtained by recognizing that the temperature at the interface depends on the local curvature of the material, as well as the speed of the front. Also the interface kinetics are highly anisotropic due to the anisotropic properties of the crystalline solid that is formed.

In a binary mixture, the interface temperature also depends on the local composition and it is possible to make a close analogy between solidification of a pure material and the approximately isothermal solidification of a supersaturated system. The basic instability of a planar or spherical front was first investigated by Mullins and Sekerka (1963, 1964), and has since been studied extensively in different contexts, for instance effects of natural and forced convection in the melt, Davis (1990).

Thus, in most practical situations, solidification interfaces undergo a fingering instability. These often develop into what is called dendrites (from the greek *dendros*, tree), see figure 8. In many metals these indeed resemble a tree with a main stem and sidebranches, where the apparently anisotropic growth is due to the anisotropy of the growth kinetics. They may typically be of the order of micrometers up to fractions of a millimeter in size. Dendrites are the most common microstructure that grows naturally during solidification of alloys and pure metals, see for instance the standard text by Kurz and

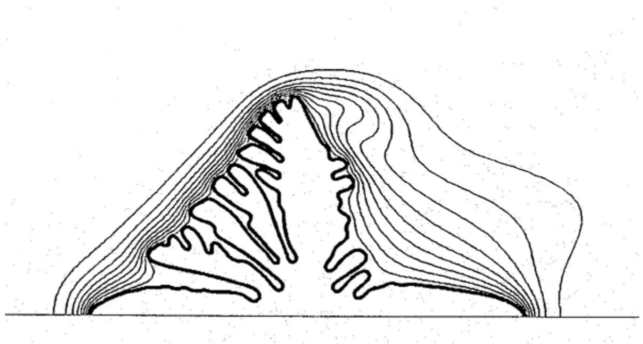

Figure 8 : Dendritic growth of a nickel nucleus in a shear flow. The innermost contour is the liquid/solid interface and the other contours are isotherms. The inflow and outflow of melt is from the left to the right.

Fisher (1992), and Huang and Glicksman (1981a,1981b), Glicksman and Marsh (1993).

In the simulation shown in figure 8 (Tönhardt and Amberg, 1998), the interface is tracked by a phase-field method. This implies that the solid/liquid interface is treated as a diffuse interface and that this is tracked by a phase-variable which is governed by a phase-field equation. The phase-field equation is derived in a thermodynamically consistent way by considering the entropy change during solidification, Wang *et al.* (1993), Fried and Gurtin (1996). This results in that the phase variable is 0 in the pure solid and 1 in the pure liquid, while it changes rapidly over the diffuse interface. The formulation of efficient phasefield models and efforts to extend their applicability and validity, is a long and continuing story, see for instance Langer (1986), Penrose and Fife (1990), Kobayashi (1991), Warren and Boettinger (1995), Karma and Rappel (1996), Karma (2001), Amberg (2003).

The simulation started from a small circular nucleus that grows into the surrounding undercooled melt, displaying the characteristic dendritic pattern with a main stem growing vertically, with secondary arms extending horizontally from the main stem. Here the orientation of the crystal lattice in the nucleus was assumed to be such that the growth is promoted in the horizontal and vertical directions.

The interesting feature that has been added in figure 8 is melt convection. We imagine that the nucleus is attached to the wall of the mold or container holding the undercooled melt, and that there is a melt flow past the wall. In keeping with the small size of the dendrite, the background flow is assumed to be a simple linear shear flow. The lower side of the domain is an insulated solid wall, the left and right side are the inflow and outflow boundaries for the fluid flow, respectively. The figure shows that the nucleus has grown into a complicated shape, a dendrite, with three main branches. Here, the fluid flow has altered the local heat transfer at the solidification front, and thus the shape of the dendrite. Due to the flow the nucleus has evolved to an asymmetric dendrite that tilts slightly to the left, upstream. Another effect of the flow is that the sidebranch growth is promoted (inhibited) on the upstream (downstream) side of the dendrite. Material properties have been chosen to approximately match those of pure Nickel, with a Prandtl number of around 0.03. A characteristic Peclet number based on the length of the vertical stem and the background velocity at this distance from the wall is around 50.

Convective effects on fully developed dendrites have not been studied using first principle simulations until quite recently. The growth of a dendrite in a shear flow was discussed above, Tönhardt and Amberg (1998, 2000a), natural convection effects have been considered by Tönhardt and Amberg (2000b). The growth of thermal dendrites in uniform forced flow has been studied by Tong et. al. (2000), Beckermann (1999a) and Diepers *et al.* (1999b), Al-Rawahi and Tryggvason (2002). Growth in a binary alloy with convection has been studied by Lan and Shih (2004). These simulations are all two dimensional, but fully three dimensional simulations of dendritic growth in a uniform forced flow has been done by Al-Rawahi and Tryggvason (2004) and Jeong, *et al.*(2001).

4 Use of Symbolic Computing to Generate FEM Code

Wherever mathematical models are used in science and engineering, one important obstacle to the development and utilization of new models is the need to produce efficient implementations of solvers for them. This is partly a question of the mathematical properties of the resulting system of equations that must be understood theoretically, but also the work involved in implementing a certain scheme is often prohibitive. In particular in an academic situation it is important to have a short turnaround time from idea to test run, in order to be able to work creatively with mathematical models.

Several tools exist that are intended to produce a working computer code from knowledge of physics and numerics in an efficient way. Out of these we may mention: diffpack[3], Ouverture[4], VECFEM[5], Matlabs PDE toolbox FEMLAB[6] and PDEase[7]. Also the more traditional commercial softwares are developing to handle more and more complex situations, or 'multi-physics'

In response to our own needs, we have developed a toolbox that uses symbolic computation to generate complete simulation codes from high level formulations of a mathematical model in the form of a system of partial differential equations, Amberg, *et al.* (1999)[8]. The entire problem definition is done in the symbolic computation application Maple. We have tried to create a toolbox that will appear natural and convenient to an applied mathematician, using only the language of applied mathematics, as implemented in the straightforward and well known Maple syntax. Spatial discretizations are done as the standard finite element method on unstructured grids. The finite elements are specified as symbolic code in a few pages of Maple procedures. Some common 1D, 2D and 3D elements are provided. The tools accommodate mixed formulations with different base functions for different variables, as often done in incompressible fluid flow. Code that is specific for a certain problem is generated within Maple, output as fortran and linked with linear algebra solvers, mesh adaptation routines, etc, to produce a rather simple standalone fortran code.

In the computational work described above on crystal growth and welding, the computations were done using codes created in this way. In addition, we may mention ongoing work on development of turbulence models for fluid flow simulations, and simulations of elastic (polymeric) liquids. In the latter case, these tools could be used in a course where students could have hands on experience of the rather complex mathematical models that are required. Other non-standard uses which might be awkward to fit into a preexisting multi-physics tool are the surfactant convection mentioned in the welding simulation above, and an ongoing implementation of a pseudospectral 3D code to simulate control of Marangoni convection.

[3] http://www.oslo.sintef.no/diffpack
[4] http://www.c3.lanl.gov/ henshaw/Overture/Overture.html
[5] http://www.uni-karlsruhe.de/ vecfem
[6] http://www.comsol.com
[7] http://www.macsyma.com/PDMain.html
[8] See also http://www.mech.kth.se/ gustava/femLego

References

Al-Rawhai, N. and Tryggvason, G. (2002): Computations of the growth of dendrites in the presence of flow. Part I-Two-dimensional Flow. *J. Comput. Phys.* vol. 180, pp. 471-496.

Al-Rawahi, N. and Tryggvason, G. (2004): Numerical simulation of dendritic solidification with convection: Three-dimensional flow. *Journal of Computational Physics*, vol. 194, pp. 677-696.

Amberg, G. (2003): A semi-sharp phase field method for quantitative phase change simulations, *Physical Review Letters*, vol. 91, pp. 265505.

Amberg, G., Tnhardt, R, and Winkler, C. (1999): Finite Element simulations using symbolic computing, *Mathematics and Computers in Simulation*, vol. 49, pp. 149-165.

Anilkumar, A. V., Grugel, R. N., Shen, X. F., Lee, C. P. and Wang, T. G. (1993): Control of thermocapillary convection in a liquid bridge by vibration, *J. Appl. Phys.*, vol. 73, pp. 4165-4170,

Bárcena, L. T., Shiomi, J. and Amberg, G. (2005): Control of oscillatory thermocapillary convection with local heating, *J. Cryst. Growth*, (submitted).

Basu, B., Enger, S, Breuer, M, and Durst, F. (2000): Three-dimensional simulation of flow and thermal field in a Czochralski melt using a block-structured finite-volume method, *J. Crystal Growth*, vol. 219, pp. 123-143.

Beckermann, C., Diepers, H.J., Steinbach, I., Karma, A., and Tong, X. (1999a): Modeling Melt Convection in Phase-Field Simulations of Solidification, *J. Computational Physics*, Vol. 154, pp. 468-496.

Benz, S., Hinz, P., Riley, R. J. and Neitzel,G. P. (1998): *J. Fluid Mech.*, vol. 359, pp. 165.

Boettinger, W.J., Coriell, S.R., Greer, A.L., Karma, A., Kurz, W., Rappaz, M. and Trivedi, R. (2000): Solidification Microstructures: Recent developments, future directions, *Acta Materialia*, vol. 48, pp. 43-70.

Cerjak, H., Bhadeshia, H.K.D.H. eds., *Mathematical Modelling of Weld Phenomena 3* (The Institute of Materials, London, 1997).

Cerjak, H., *Mathematical Modelling of Weld Phenomena 6* (The Institute of Materials, London, 2001).

Chang, C.E. & Wilcox, W.R. (1976): Analysis of surface tension driven flow in floating zone melting, *Intl J. Heat Mass Transfer*, vol. 19, pp. 355-356.

Chatterjee, A., Prasad, V. and Sun, D. (2000): A full 3-dimensional adaptive finite volume scheme for transport and phase-change processes, part II: Application to crystal growth, *Numerical Heat Transfer Part A - Applications*, vol. 37, pp. 823-843.

Chen, G., Lizee, A., & Roux, B. (1997): Bifurcation analysis of the thermocapillary convection in cylindrical liquid bridges. *Journal of Crystal Growth*, vol. 180, pp. 638-647.

Chun, C. -H. and Wuest, W. (1979): *Acta Astron.*, vol. 6, pp. 1073.

Cröll, A., Müller-Sebert, W., Benz, K.W. and Nitsche, R. (1991): *Microgravity Sci. Technol.*, III/4, pp. 204.

Cröll., Szofran, F. R., Dold, P., Benz, K. W. and Lehoczky, S. L. (1998): Floatingzone growth of silicon in magnetic fields 2. strong static axial fields. , *J. Crystal Growth*, vol. 183, pp. 554-563 .

David, S.A. and Vitek, J.M. eds., *International Trends in Welding Science and Technology* (ASM International, Materials Park, OH, 1993).

Davis, S.H. (1990): Hydrodynamic interactions in directional solidification, *J. Fluid Mech.*, vol. 212, pp. 241-262.

Diepers, H.J., Beckermann, C., and Steinbach, I. (1999b): Simulation of Convection and Ripening in a Binary Alloy Mush Using the Phase-Field Method, *Acta Materialia*, Vol. 47, pp. 3663-3678.

Do-Quang, M., Melt Convection in Welding and Crystal Growth, Doctoral thesis, Dept of Mechanics, KTH, S-100 44 Stockholm, Sweden, (2004).

Do-Quang, M., Amberg, G. and Pettersson, C.-O. (2005): Experimental and numerical study of the influence of sulfur redistribution in welding of SAF-2507 stainless steel, Science and Technology of Welding and Joining, submitted.

Dornberger, E., Tomzig, E., Seidl, A., Schmitt, S., Leister, H.-J., Schmitt, Ch. and Müller, G. (1997): Thermal simulation of the Czochralski silicon growth process by three different models and comparison with experimental results, *J. Crystal Growth*, vol. 180, pp. 461-467.

Dressler, R. F. and Sivakumaran, N. S. (1988): Non-contaminating method to reduce Marangoni convection in microgravity float zones, *J. Crystal Growth*, vol. 88, pp. 148-158.

Enger, S, Basu, B., Breuer, M, and Durst, F. (2000): Numerical study of three-dimensional mixed convection due to buoyancy and centrifugal force in an oxide melt for Czochralski growth., *J. Crystal Growth*, vol. 219, pp. 144-164.

Evstratov, I.Yu., Kalaev, V.V., Zhmakin, A.I., Makarov, Yu.N., Abramov, A.G., Ivanov, N.G., Smirnov, E.M., Dornberger, E., Virbulis, J., Tomzig, E., v.Ammon, W. (2000): Modeling analysis of unsteady three-dimensional turbulent melt flow during Czochralski growth of Si crystals, *3rd International Workshop on Modeling Crystal Growth* - October 18-20 2000, Hauppauge, New York USA.

Fainberg, J., Leister, H.-J. and Müller, G., Numerical simulation of the LEC-growth of GaAs crystals with account of high-pressure gas convection, *J. Crystal Growth*, vol. 180, pp. 517-523.

Fried, E., Gurtin, M. E. (1997): A phase-field theory for solidification based on a general anisotropic sharp-interface theory with interfacial energy and entropy, *Phys. D*, 91, pp. 143-181, (1996).

Glicksman, M.E. and Marsh, S.P. (1993): The Dendrite, in: D.T.J. Hurle (Ed.), *Handb. Cryst. Growth*, vol.1, ch.13, Elsevier, Amsterdam, pp. 1077-1122.

Huang, S.-C. and Glicksman, M.E. (1981a): Fundamentals of dendritic solidification. I. Steady-state tip growth. *Acta Met.*, vol. 29 no.5, pp. 701-716.

Huang, S.-C. and Glicksman, M.E. (1981b): Fundamentals of dendritic solidification. II. Development of sidebranch structures. *Acta Met.*, vol. 29 no. 5, pp. 717-734.

Jeong J.-H., Goldenfeld, N. and Dantzig, J.A. (2001): Phase field model for three-dimensional dendritic growth with fluid flow, *Physical Review E*, vol. 64, pp. 041602.

Jing, C.J., Imaishi, N., Sato, T. and Miyazawa, Y. (2000): Three-dimensional numerical simulation of oxide melt flow in Czochralski configuration, *J. Crystal Growth*, vol. 216, pp. 372-388.

Kaiser, T. and Benz, K.W. (1998): Floating-zone growth of silicon in magnetic fields III. Numerical Simulation, *J. Crystal Growth*, vol. 183, pp. 564-572.

Kaiser, T., Magnetohydrodynamische Effekte bei der Halbleiterkristallzüchtung, Thesis, Albert-Ludwigs-

Universitt Freiburg, Shaker Verlag (1998).

Kamotani, Y., Lee, J. and Ostrach, S. (1992): An experimental study of oscillatory thermocapillary convection in cylindrical containers., *Phys. Fluids A*. vol. 4, pp. 955-962.

Kamotani, Y. (1997): Surface tension driven convection in microgravity , *Advances in the Astronautical Sciences*. Vol. 96, pp. 487-499.

Kamotani, Y., Ostrach, S. & Pline, A. (1998): Some temperature field results from the thermocapillary flow experiment aboard USML-2 spacelab, *Advances in Space Research*. vol. 22, pp. 1189-1195.

Kamotani, Y., Ostrach, S. and Masud, J. (2000): Microgravity experiments and analysis of oscillatory thermocapillary flows in cylindrical containers. *J. Fluid Mech.*, vol. 410, pp. 211-233.

Karma, A. (2001): *Phys. Rev. Lett.* vol. 87, pp. 115701.

Karma, A. and Rappel, W.-J. (1996): Phase-field method for computationally efficient modeling of solidification with arbitrary interface kinetics, *Physical Review E*, vol. 53, pp. R3017-R3020.

Kawamura, H., Ueno, I., Tanaka, S. and Nagano, D., Osillatory, Chaotic and Turbulent Thermocapillary Convections in a Half-Zone Liquid Bridge, Proc of Turbulent Shear Flows and Phenomena 2, Stockholm, June 2001.

Kimura, H., Harvey, M. F., Ofconner, D. J., Robertson, G. D. and Valley, G. C. (1983):, Magnetic field effects on floating-zone Si crystal growth, *J. Crystal Growth*, vol. 62, pp. 523-531.

Kobayashi, R. (1991): *Bull. Jpn. Soc. Ind. Appl. Math.*, vol. 1, pp. 22.

Kurz, W. and Fisher, D.J., *Fundamentals of Solidification*, Trans Tech Publications (1992)

Lan, C.W., Chian, C.H. and Tu, C.Y., Three-dimensional Simulation of Floating-Zone Crystal Growth, The 2000 CIChE Annual Meeting and Conferences, Proceeding 2000 Symposium on Transport Phenomena and Applications, pp. 1-6 (2000).

Lan, C. W. and Shih, C. J. (2004): Efficient phase field simulation of a binary dendritic growth in a forced flow, *Phys. Rev. E* vol. 69, pp. 031601.

Lan, C. W. and Yeh, B. C. (2005): Effect of rotation on heat flow, segregation, and zone shape in a small-scale floating-zone silicon growth under axial and transversal magnetic field, *FDHP*, vol. 1, no. 1.

Langer, J.S., Models of pattern formation in first-order phase transitions Directions in condensed matter physics. *Memorial volume in honor of Sheng-keng Ma*. World Scientific, Singapore, xiv+252, pp.165-186, (1986).

Langlois, W.E. (1985): Buoyancy-driven flows in crystal-growth melts, *Ann. Rev. Fluid Mech.*, vol. 17, pp. 191-215.

Lappa, M. (2003): Three-dimensional numerical simulation of Marangoni flow instabilities in floating zones laterally heated by an equatorial ring, *Physics of Fluids*, vol. 15, no. 3, pp. 776-789.

M. Lappa (2004a): *Fluids, Materials and Microgravity: Numerical Techniques and Insights into the Physics*, Elsevier Science (Oxford, 2004), 538pp. - ISBN 00-804-4508-X.

Lappa, M. (2004b): Combined effect of volume and gravity on the three-dimensional flow instability in non-cylindrical floating zones heated by an equatorial ring, *Physics of Fluids*, vol. 16, no. 2, pp. 331-343.

Lappa, M. (2005a): On the nature and structure of possible three-dimensional steady flows in closed and open parallelepipedic and cubical containers under different heating conditions and driving forces, *FDMP*, vol. 1, no. 1, pp. 1-19.

Lappa, M. (2005b): Thermal convection and related instabilities in models of crystal growth from the melt on earth and in microgravity: Past history and current status, *Cryst. Res. Technol.*, vol. 40, no. 6, pp. 531-549.

Lee, C. P. (1998): The balancing of thermocapillary flow in a floating zone by ripple-driven streaming, *Physics of Fluids*, vol. 11, pp. 2765-2780.

Lee, C. P., Anilkumar, A. V. and Wang, T. G. (1996): Streaming generated in a liquid bridge due to nonlinear oscillations driven by the vibration of an endwall, *Physics of Fluids*, vol. 8, pp. 3234-3246.

Leon, N. D., Guldberg, J. and Salling, J. (1981): Growth of homogeneous high resistivity FZ silicon crystals under magnetic field bias, *J. Crystal Growth*, vol. 55, pp. 406-408.

Levenstam, M., Amberg, G., Carlberg, T. and Andersson, M. (1996): Experimental and numerical studies of thermocapillary convection in a floating zone like configuration, *J. Crystal Growth*, vol. 158, pp. 224-230.

Levenstam, M., Amberg, G. and Winkler, C. (2001): Instabilities of thermocapillary convection in a half-zone at intermediate Prandtl numbers, *Physics of Fluids*, vol. 13, pp. 807-816.

Lipchin, A. and Brown, R.A. (1999): Comparison of three turbulence models for simulations of melt convection in Czochralski crystal growth of silicon, *J. Crystal Growth*, vol. 205, pp. 71-91.

Müller, G. and Rupp, R. (1991): The role of Marangoni convection in the growth of GaAs crystals by the floating zone technique under microgravity, *Crystal Properties and Preparation*, vol. 35, pp. 138-154.

Mullins, W.W. and Sekerka, R.F. (1963): Morphological Stability of a Particle Growing by Diffusion or Heat Flow, *Journal of Applied Physics*, Vol. 34, pp. 323-329.

Mullins, W.W. and Sekerka, R.F. (1964): Stability of a Planar Interface During Solidification of a Dilute Binary Alloy, *Journal of Applied Physics*, Vol. 35, pp. 444-451.

Mundra, K. and Debroy, T., (1993a): Calculation of Weld Metal Composition Change in High-Power Conduction Mode Carbon Dioxide Laser-Welded Stainless Steels, *Metallurgical Transactions B*, vol. 24B, pp. 145-155.

Mundra, K. and Debroy, T. (1993b): Toward Understanding Alloying Element Vaporization during Laser Beam Welding of Stainless Steel, *Welding Journal Research Supplement* vol. 72, pp. 1-9.

Neitzel, G. P., Chang, K. T., Jankowski, D. F. & Mittelmann, H. D. (1993): Linear stability theory of thermocapillary convection in a model of the float-zone crystal-growth process., *Physics of Fluids A*, vol. 5, pp. 108-114.

Neitzel, G. P., Law, C.C., Jankowski, D. F. & Mittelmann, H. D. (1991): Energy stability of thermocapillary convection in a model of the float-zone crystal-growth process. ii: Nonaxisymmetric disturbances, *Physics of Fluids A*, vol. 3, pp. 2841-2846.

Oreper, G.M. and Szekely, J. (1984): Heat- and Fluid-Flow Phenomena in Weld Pools, *Journal of Fluid Mechanics*, vol. 147, pp. 53-79.

Penrose, O. and Fife, P. (1990): *Physica (Amsterdam)*, vol. 43D, pp. 44.

Petrov, V., Schatz, M. F., Muehlner, K. A., VanHook, S. J., McCormick, W. D., Swift, J. B. and Swinney, H. L. (1996): Model-independent nonlinear control algorithm with application to a liquid bridge experiment., *Phys. Rev. Lett.*, vol. 77, pp. 3779-3782.

Petrov, V., Muehlner, K. A., VanHook, S. J. and Swinney, H. L. (1998): Model-independent nonlinear control algorithm with application to a liquid bridge experiment., *Phys. Rev. E*, vol. 58, pp. 427-433.

Preisser, F., Schwabe, D., & Scharmann, A. (1983): Steady and oscillatory thermocapillary convection in liquid columns with free cylindrical surface *J. Fluid Mech.*, vol. 126, pp. 545-567.

Rao, M.M. and Shyy, W. (1997): Moving boundary computation f the float-zone process, *Int. J. Num. Meth. Eng.*, vol. 40, pp. 1231-1261.

Ratnieks, G., Muiznieks, A., Buligins, L., Raming, G., Mühlbauer, Lüdge ad Riemann,H. (2000): influence of the three dimensionality of the HF electromagnetic field of resistivity variations in Si single crystals during FZ growth, *J. Crystal Growth*, vol. 216, pp. 204-219.

Robertson, G. D. and OfConnor, D. J. (1996a): Magnetic field effects on float-zone si crystal growth : Ii. strong transverse fields, *J. Crystal Growth*, vol. 76, pp. 100-110.

Robertson, G. D. and OfConnor, D. J. (1986b): Magnetic field effects on float-zone si crystal growth : Ii?. strong axial fields, *J. Crystal Growth*, vol. 76, pp. 111-122.

Rupp, R., Müller, G., & Neumann, G. (1989): Three-dimensional time dependent modelling of the Marangoni convection in zone melting configurations for GaAs, *J. Cryst. Growth*, vol. 97, pp. 34-41

Schatz, M. F. and Neitzel, G. P. (2001): Experiments on thermocapillary instabilities, *Annu. Rev. Fluid Mech.*, vol. 33, pp. 93-127.

Scheel, H.J. (2000): Historical aspects of crystal growth technology, *Journal of Crystal Growth*, vol. 211, pp. 1-12.

Schwabe, D. and Scharmann, A. (1979): *J. Cryst. Growth*, vol. 46, pp. 125.

Shen, Y., Neitzel, G.P., Jankowski, D.F., & Mittelman, H.D. (1990): Energy stability of thermocapillary convection in a model of the float-zone crystal-growth process, *J. Fluid Mech.*, vol. 217, pp. 639-660.

Shen, X. F., Anilkumar, A. V., Grugel, R. N. and Wang, T. G. (1996): Utilizing vibration to promote microstructural homogeneity during floating-zone crystal growth processing , *J. Cryst. Growth*, vol. 165, pp. 438-446.

Shiomi, J., Amberg, G. and Alfredsson, P.H. (2001):

Active control of oscillatory thermocapillary convection, *Phys. Rev. E*, vol. 64, pp. 031205.

Shiomi, J. and Amberg, G. (2002): Active control of a global thermocapillary instability , *Phys. Fluids*, vol. 14, pp. 3039-3045.

Shiomi, J., Kudo, M., Ueno, I., Kawamura, H. and Amberg, G. (2003): Feedback control of oscillatory thermocapillary convection in a half-zone liquid bridge, *J. Fluid Mech.*, vol. 496, pp. 193-211.

Shiomi, J. and Amberg, G. (2005): Proportional control of oscillatory thermocapillary convection in a toy model, *Eur. J. Mech. B/Fluids*, vol. 24 pp296-313.

Shiomi, J. and Amberg, G. (2005): Numerical investigation of feedback control of thermocapillary instability, *Phys. Fluids*, vol. 17, pp. 1-12.

Smith, M. K., & Davis, S. H. (1983): Instabilities of dynamic thermocapillary liquid layers Part 1. Convective instabilities *J. Fluid Mech.*, vol. 132, pp. 119-144.

Tong, X., Beckermann, C., and Karma, A. (2000): Velocity and Shape Selection of Dendritic Crystals in a Forced Flow, *Physical Review E*, Vol. 61, pp. R49-R52.

Tönhardt, R, and Amberg, G. (2000a): Dendritic growth of randomly oriented nuclei in a shear flow, *Journal of Crystal Growth*, vol. 213, pp. 161-187.

Tönhardt, R, and Amberg, G. (2000b): Simulation of natural convection effects on SCN crystals, *Physical Review E*, vol. 62, pp828-836.

Tönhardt, R. and Amberg, G. (1998): Phasefield Simulation of Dendritic growth in a Shear Flow, *J. Crystal Growth*, vol. 194, pp. 406-425.

Tsukada T., Kobayashi H., Jing C. J., Imaishi N. (2005): Numerical simulation of CZ crystal growth of oxide, *FDNP*, vol. 1.

Ueno, I., Ono, Y., Nagano, D., Tanaka, S. and Kawamura, H., Modal oscillatory structure and dynamic particle accumulation in liquid-bridge Marangoni convection, Proc of the 4th JSME-KSME Thermal Engineering Conference, October 1-6, 2000, Kobe, Japan., vol. 3, pp. 265-270, (2000).

Velten, R., Schwabe, D. and Scharmann, A. (1991): The periodic instability of thermocapillary convection in liquid bridges, *Physics of Fluids*, vol. 3A, pp267-279.

Wang, S.-L., Sekerka, R.F., Wheeler, A.A., Murray, B.T., Coriell, S.R., Braun, R.J. and McFadden, G.B. (1993): Thermodynamically-consistent phase-field models for solidification, *Phys. D*, vol.69, no.1-2, pp.189-200.

Wanschura, M., Shevtsova, V. M., Kuhlmann, H. C. & Rath, H. J. (1995): Convective instability mechanisms in thermocapillary liquid bridges, *Phys. Fluids*, vol. 7, pp. 912-925.

Warren, J.A., and Boettinger, W.J. (1995): *Acta Metall. Mater.*, vol. 43, pp. 689-703.

Winkler, C., Amberg, G., Inoue, H., and Koseki, T. A Numerical and Experimental Investigation of Qualitatively Different Weld Pool Shapes, in H. Cerjak and H.K.D.H. Bhadeshia eds, *Mathematical Modelling of Weld Phenomena 4*, (The Institute of Materials, London, 1998)

Winkler, C., Amberg, G., Inoue, H., Fuji, M. and Koseki, T. The role of surfactant mass transfer in GTA-welding, to appear in H. Cerjak and H.K.D.H. Bhadeshia eds, *Mathematical Modelling of Weld Phenomena 5*, (The Institute of Materials, London, 2000a)

Winkler, C., Amberg, G., Inoue, H., Koseki, T. and Fuji, M. (2000b): The effect of surfactant redistribution on the weld pool shape during GTA-welding, *Science and Technology of Welding and Joining*, vol. 5, pp. 1-13.

Xiao, Q. and Derby, J.J. (1994): Heat transfer and interface inversion during the Czochralski growth of yttrium aluminum garnet and gadolinium gallium garnet, *J. Crystal Growth*, vol. 139, pp. 147-157.

Xu, J-J., & Davis, S. H. (1984): Convective thermocapillary instabilities in liquid bridges. *Physics of Fluids* , vol. 27, pp. 1102-1107.

Smith, M. K. (1986): Instability mechanisms in dynamic thermocapillary liquid layers. *Physics of Fluids*, vol. 29, pp. 3182-3186.

Zou, Y.F., Wang, G.X., Zhang, H., Prasad, V. and Bliss, D.F. (1997): Macro-segregation, dynamics of interface and stresses in high pressure LEC grown crystals, *J. Crystal Growth*, vol. 180, pp. 517-523.