

FLUID FLOWS IN CRYSTAL GROWTH AND OTHER MATERIALS PROCESSES

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ABSTRACT

A few problem areas in materials science are reviewed with regard to the importance of fluid flows, in particular flow instabilities and turbulence. One class of such flows in melts are driven by surface tension gradients caused by temperature differences. These are frequently responsible for instabilities or turbulence that is often undesired in practical processes, as in crystal growth. Another area that is of obvious importance in materials processes is solidification. Fluid flows are crucial here, since the growth rate, the shape and size of the solid structures are greatly influenced by melt convection.

THERMOCAPILLARY CONVECTION

Thermocapillary convection becomes important whenever a strong temperature gradient is present over a free liquid surface, in particular if the surface tension is strong. Also, if the dimensions of the liquid volume is 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.

The basic phenomenon of thermocapillary convection is understood by considering a small section of a free liquid surface with a temperature dependent surface tension,

$$\sigma = \sigma_0 + \gamma \cdot (T - T_0), \quad (1)$$

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 differ-

ence in surface tension must be balanced by a shear stress in the fluid. The typical 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 viscosity.

The above would typically be true for pure fluids. In the presence of a possibly surface active additive, concentration gradients would drive a *solutocapillary* 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). With a decreased (increased) concentration the surface tension increases (decreases), and a restoring force thus 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.

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, highly uniform properties. This is presently a large industry of obvious and growing importance for electronics, optics, laser technology and other applications, with the largest area being semiconductors accounting for 60% of the 20000 tons produced in 1999, Scheel (2000). One possible future application area would be high temperature superconductors, the succes-

ful commercialization of which would be of immense economic importance.

There are many different processes that are used, see Scheel (2000) for a current 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 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 crystals, but it is complicated by the fact that gravity may have a large influence on the suspended melt drop.

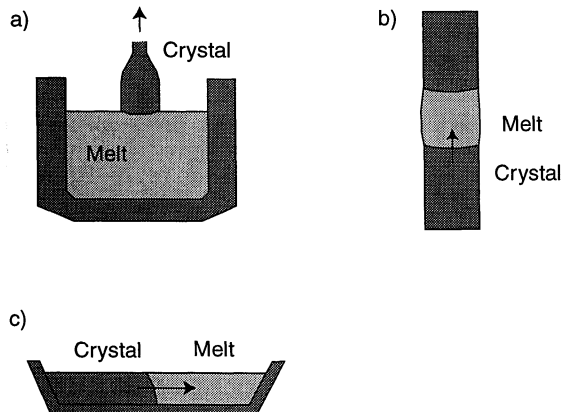


Figure 1: Sketches the geometrical configurations in a) Czochralski growth, b) floating zone, c) Horizontal Bridgman growth.

Common features of these methods are the presence of a free surface and very high temperature gradients. This is thus 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) and numerous studies have since then investigated different aspects of thermocapil-

lary convection in configurations resembling crystal growth processes.

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 maintaining a temperature difference between the two rods causing a temperature gradient, and hence a surface tension gradient, along the free surface. The main objective of most studies relating 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 float-zone method typically have periodic axial variations in dopant concentration, so called striations, which are attributed to an oscillatory thermocapillary convection in the melt. The fluid mechanical problem that presents itself is thus to understand the flow instabilities that lead to unsteady motion.

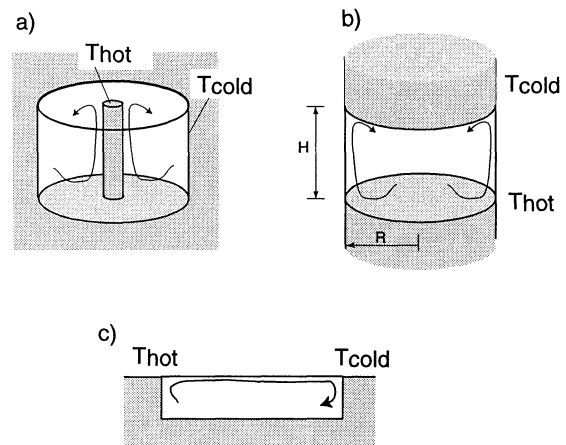


Figure 2: Sketches of a few model problems related to crystal growth a) Annular configuration, b) Half zone, c) Horizontal rectangular cavity.

The fundamental instability mechanisms in thermocapillary 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. (1991,1993), Wanschura et.al. (1995), Chen et al. (1997) and Levenstam et al (2001). Full numerical simulations of the developed instability has been made by, among others, Rupp et al(1989), Lev-

enstam and Amberg (1995) and Levenstam et al (2001).

Experimentally the half zone has been a popular model geometry, starting with Preisser et.al. (1983). See also the recent review by Schatz and Neitzel (2001). It is quite difficult to do well controlled experiments using technically interesting fluids with small Prandtl numbers such as semiconductor melts. Instead a large literature has appeared that study thermocapillary flows using fluids with Prandtl numbers greater than one, typically silicon 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 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.

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 et al, 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, 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

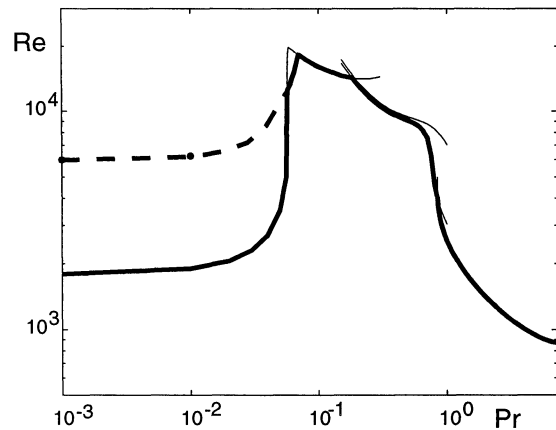


Figure 3: Stability of the flow in a half zone. Below the lower curve the flow is steady and axisymmetric. At Prandtl numbers above 1, the flow is oscillatory above the curve. At low Prandtl numbers the flow is steady and 3D between the full and the dashed line, and oscillatory above the dashed line.

range, with critical Reynolds numbers around ten times larger than the levels outside. This is due to the 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. In the half zone this has been attempted by Petrov et al (1996) who applied a non-linear control algorithm using a local temperature measurement close to the free surface and heating a thermoelectric element placed at a location diametrically opposite the measurement. The oscillations could be suppressed at the sensor for Marangoni numbers 1.3 times the critical value, however infrared visualization revealed the presence of standing waves with antinodes at the feedback element and the sensor.

Practical float zone processes and experiments are (naturally) more complicated than the half zone discussed above. The tempera-

ture differences that are used are such as to give thermocapillary Reynolds numbers in 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 speed that is apparently chaotic in time. Model experiments that investigate the chaotic regime in a systematic fashion have appeared only recently, Ueno et.al. (2000), Kawamura et.al. (2001), who manage to make 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.

Notice that the thermocapillary Reynolds number actually overestimates the *actual* Reynolds number in the flow. Typically the product of actual maximum velocity and diameter divided by kinematic viscosity, would scale as $Re^{2/3}$ giving much more modest values. A reasonable comparison could be made with a driven cavity at $Re=1000-10000$. The flow in a real float zone is thus highly chaotic, even if the Reynolds number is hardly high enough for engineering turbulence models to give good results, Kaiser (1998), regardless of all the well known difficulties with separation and recirculating flows, let alone wall boundary conditions or wall corrections for a thermocapillary boundary, etc.

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 considered as the most severe. In order to stabilize the flow, differential rotation of the rods, magnetic fields etc, may be applied. 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.

The annular configuration.

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 et.al. (1997,1998,2000) investigated the onset of oscillations in this geometry using silicone oil with Prandtl number around 27.

The group of Kamotani and Ostrach have argued that free surface deformations are an intrinsic part of the instability mechanism, both in the annular geometry and in the half zone (Masud et.al., 1997), and have proposed a mechanism for the instability that relies on the coupling between the thermocapillary heat transfer and a time dependent surface deformation. However, this explanation is not without problems. The first observation is that the time dependent surface deformations that are observed during oscillations are very small, in the order of microns for a typical experiment in silicone oil, Kamotani et. al. (2000). Furthermore, there have been several reasonable comparisons between experiments and stability in the half zone case that assume an undeformed free surface, Wanschura et.al. (1995), Neitzel et.al. (1993), Levenstam et.al. (2001). We are not aware of any published direct comparisons between stability theory and experiments for the annular geometry except for Lavalley (1997) and Lavalley et. al. (2001), which show good agreement between experiments and simulation with an undeformed free surface.

Schwabe et. al. (1992) studied experimentally the mode shape as a function of aspect ratio for very shallow annular containers. They observed a selection of azimuthal wavenumbers that was similar to what has been observed earlier in the half zone case. In very shallow containers, $H/R = 0.013$, azimuthal wavenumbers as high as 22 were obtained.

An attempt to introduce an active feedback control has been made by Shiomi et al (2001), who introduced a local heating of the horizontal surface, based on a temperature measurement taken at a different position. As in the control experiment by Petrov et.al. (1996), control is successful in suppressing oscillation at the sensor location. However to completely eliminate oscillations, the temperature must be

measured at several locations.

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 size 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 these elevated 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). 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 $k-\epsilon$ model. As they show, the melt flow in Czochralski growth has many of the features that are notoriously difficult to treat using $k-\epsilon$ 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).

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 and Bhadeshia (1997, 1998), David and Vitek (1993)).

The molten weld pool develops directly beneath the electrode when the current is turned on and its shape and size is highly influenced by the heat and fluid flow in the molten zone. The heat and fluid flow affect the temperature distribution and thus determine the quality of the solidified weld fusion zone.

The fluid flow in the weld pool is mainly driven by forces due to surface tension gradients (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 modelling 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 recent research, physicochemical phenomena, such as adsorption and desorption to the surface, are considered as important for realistic modelling, Winkler et.al. (1998, 2000).

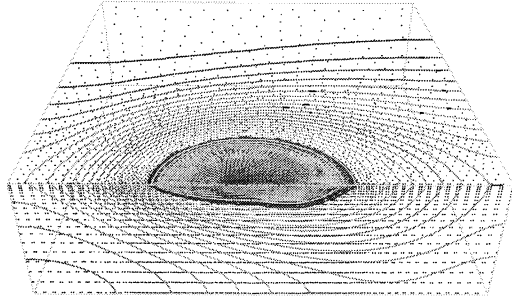


Figure 4: Isotherms and flow vectors on the surface and symmetry plane of a plate during welding.

Figure 4 shows stationary temperature and velocity distributions for a 3D GTA-welding simulation. The electrode is moving with a constant speed of $0.5m/min$ 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. Isotherms are shown on the upper surface of the plate and on the vertical symmetry plane oriented along the travelling direction of the electrode. In these planes also the velocity fields are plotted. The solidification front of the weld pool where liquid material suddenly changes to solid is shown as a surface bounding the weldpool from below. It is observed that 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, resulting in this case in a total of 6 rotating vortex motions influencing the weld pool depth and width. Moreover, the velocity field at the surface of the specimen determines the streamlines defining the travelling paths of, for example, slag particles.

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, is 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 paramount in determining the structure of the material, thus making this an area of important applications that should interest fluid mechanists.

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 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 in fact 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 temper-

ature 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 5. 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 micrometers up to fractions of a millimeter in size. Dendrites are maybe the most common microstructure that grows naturally during solidification of alloys and pure metals, see for instance the standard text by Kurz and Fisher (1992), and Huang and Glicksman (1981a,1981b), and Glicksman and Marsh (1993).

In the simulation shown in figure 5 (Tönhardt and Amberg, 1998), the interface is tracked by a phase-field method, Langer (1986), Kobayashi (1991), Warren and Boettinger (1995), Karma and Rappel (1996). 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.

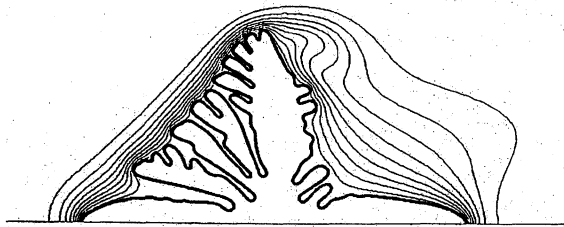


Figure 5: Dendritic growth of a nickel nucleus in a shear flow. The innermost contour is the liquid/solid interface and the other contours are isotherms. The inflow and outflow of melt is from the left to the right.

The simulation started from a small circu-

lar 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 5 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 dendrites in uniform forced flow has been studied by Tong et. al. (2000), Beckermann (1999a) and Diepers et.al. (1999b). These simulations are all two dimensional, but a fully three dimensional simulation of dendritic growth in a uniform forced flow has been done by Al-Rawahi and Tryggvason (2000).

Modeling of casting, the mushy zone

As these dendrites continue to grow, they may after some time occupy a macroscopic region and can then be treated as a porous material made up of solid crystals bathed in residual melt. For instance in a casting, where

a molten alloy is solidified by cooling the walls of the mould, dendrites usually grow from the walls into the melt. This effectively porous region is referred to as a 'mush'. The speed at which this mush advances is in principle determined by the conditions at the edge of the mush, i.e. the local heat and mass transfer at the tips of the dendrites that constitute the mush edge, i.e. the interaction between dendritic growth and flow that is addressed above. Another important class of phenomena relate to the mass transfer inside the mush. Due to the thermodynamics of phase change, the melt in the mush is typically enriched in alloying elements. This induces density gradients and thus convective flow.

The effects on the final solid of such convective motions (see Huppert, 1990) go by many different names in materials science. In the directional casting of single crystal turbine blades, the fossil traces of a convecting plume in the mush as called a 'freckle', and is highly detrimental, Worster (1992,1997), Amberg and Homsy (1993).

In large scale castings the enriched melt in the mushy region may slowly convect through the porous mush to the top or the bottom of the cast, as the case may be, causing accumulation of light element at the top and heavy at the bottom of the cast. Such macrosegregations can now be reasonably modelled, also for complex systems, Schneider and Beckermann (1995a,1995b). In the continuous casting of steel there are many important phenomena related to the use of strong magnetic fields to control the melt flow, Davidson (1999).

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