Stability of Numerical Simulations of Dendritic Solidification*

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Novel developments in the numerical simulation of diffusion limited dendritic growth are discussed in the context of two-dimensional simulations of solidification of pure substances and of binary alloys. The three most important numerical difficulties encountered in the simulations of dendritic growth of binary alloys are discussed: 1) the need to accurately calculate the position and velocity of the interface as part of the solution; 2) the disparity of length scales between the thermal diffusion length and the solute diffusion length; and 3) the instability of the solid-liquid interface, particularly at high concentrations of solute. Dealing with the third difficulty constitutes the main objective of this paper. The stability of calculations is studied using the continuity condition on the heat flux across the interface and numerical simulations. The latter are used to assess the current modeling capabilities and the hurdles faced to produce more powerful simulators.

Key Words: Interface Stability, Finite Elements, Binary Alloys, Interface Tracking

1. Introduction

Directional solidification of alloys is used in the manufacturing of turbine blades for high temperature gas turbines⁽¹⁾. In this process it is crucial to guarantee a product free of defects such as macrosegregation and to be able to control the microstructure of the final castings to obtain optimal creep and fatigue properties. Numerical models have become extremely useful in recent years to analyze and understand the basic physical processes involved in the production of the castings⁽²⁾. However, models for the prediction of dendritic structure are still not able to do a satisfactory job. The main difficulty is the need to properly model the developing solid-liquid interface.

The numerical simulation of dendritic growth in solidification processes is notoriously difficult due to three main factors that have a direct impact in the numerical algorithms. First, the position and velocity

of the solid-liquid interface must be computed as part of the solution, which makes the problem highly non-linear and requires the tracking of a continuous motion using discrete steps. Second, the problem involves a number of different length scales that range over several orders of magnitude; specifically in alloys the thermal diffusion length and the solute diffusion length differ by four orders of magnitude. The latter forces the modeler to use very fine computational meshes over large spatial regions. The third reason is the inherent instability of the solid-liquid interface, which also makes it very hard to achieve numerical stability. This is the focus of attention of this article, and will be discussed in section 3.

In order to make the problem more tractable, many researchers have tried to avoid calculating the position of the interface by reformulating the problem; this has produced a variety of methods such as the enthalpy method, the level-set method and the phase-field method^{(3)-(5)*1}, in which the position of the interface is recovered approximately from the value

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*1 Due to the large amount of literature in the subject, no attempt is made here to make the references list exhaustive.

of a field variable that is computed instead. Of these methods, the most successful one has been the phase-field method, which has been used extensively to calculate dendritic structures in pure materials and to a limited extent in binary alloys⁽⁶⁾⁻⁽⁸⁾. This method; however, is difficult to formulate and use; in particular, it is not easy to relate the parameters in the phase-field variable to the physical parameters.

In simulations of solidification of binary alloys, to avoid the difficulties associated with the disparity between the thermal diffusion length and the solutal diffusion length scale, the approach has been to assume that the temperature field is known and does not need to be calculated as part of the solution⁽⁸⁾⁻⁽¹²⁾, simplifying the simulations to the solution of the solute-concentration only. Finally, to be able to deal more effectively with the instability of the interface in the solidification of binary alloys, the simulations have been restricted to very dilute alloys^{(9),(12)-(15)} or to solidification of eutectics^{(7),(16)}.

The first simulations that include the full model, calculating the temperature and solute concentration fields over the full hypoeutectic range, are reported in Ref.(17), which is used as the basis for the present work. The numerical model was developed based on the ideas contained in Refs.(18)-(20). It solves the energy equation in a fixed mesh of bilinear quadrilateral finite elements; the interface is tracked using marker points that move according to the interface velocity, and whose number changes to maintain proper resolution as the interface evolves. The solute-concentration in the liquid is solved independently on a variable mesh of quadratic triangular elements that is generated anew at every time step to account for the geometric changes in the domain, the thermal and solutal concentration fields are coupled at the solid-liquid interface. The details of the numerical algorithm are given in Refs.(17), (20).

2. Mathematical Model

It is assumed that the solidification/melting process is diffusion limited; most of the material properties are different but constant in each phase; the latent heat is constant and the densities are equal in the solid and liquid, so fluid flow due to shrinkage and natural convection is ignored. The energy equation is given by

$$\rho c_{PS} \frac{\partial T_S}{\partial t} = \kappa_S \nabla^2 T_S \quad \text{in the solid} \quad (1.a)$$

and

$$\rho c_{PL} \frac{\partial T_L}{\partial t} = \kappa_L \nabla^2 T_L \quad \text{in the liquid} \quad (1.b)$$

that at the solid-liquid interface must satisfy the conditions

$$T_S = T_L = T_I \quad (2.a)$$

and

$$(\kappa_S \nabla T_S - \kappa_L \nabla T_L) \cdot \hat{n} = \rho [L + (c_{PL} - c_{PS})(T_I - T_m)] V \quad (2.b)$$

In the above equations, the subscripts *S* and *L* denote the solid and liquid phases, respectively; ρ is the density; c_P is the specific heat; κ is the thermal conductivity; $\hat{n}$ is the unit vector normal to the interface pointing into the liquid; L is the latent heat; T_I and T_m are the local interface and equilibrium liquidus temperature, respectively; and V is the local normal interface velocity.

Because in the metallic alloys of our interest the solute diffusion in the solid is several orders of magnitude smaller than in the liquid, solute diffusion in the solid is neglected; the solute concentration is solved only in the liquid and is given by

$$\frac{\partial C_L}{\partial t} = D_L \nabla^2 C_L \quad (3)$$

with interface conditions before eutectic solidification

$$(-D_L \nabla C_L) \cdot \hat{n} = (1 - k) C_L V \quad (4.a)$$

and at eutectic solidification

$$C_L = C_E \quad (4.b)$$

In Eqs.(3), (4.a) and (4.b) D_L is the solutal diffusion coefficient in the liquid, k is the equilibrium partition ratio and C_E is the eutectic concentration.

The local interface temperature is given by the generalized Gibbs-Thompson relation⁽¹⁹⁾

$$T_I - T_m + \frac{\gamma T_m}{L \rho} \mathcal{C} + \frac{V}{v} + \frac{T_m (c_{PL} - c_{PS})}{L} \times \left[T_I \ln \left(\frac{T_I}{T_m} \right) + T_m - T_I \right] = 0 \quad (5)$$

where γ is the surface energy; $\mathcal{C}$ is the local interface curvature, and v is the kinetic mobility. The liquidus of the phase diagram is approximated linearly by

$$T_m = T_{m0} + m C_L \quad (6)$$

where m is the slope of the liquidus line and T_{m0} is the melting point of the pure solvent.

The equations are solved using the finite element method with a standard Galerkin weighted residuals formulation. The solution of the energy equation is carried out on a fixed mesh of bilinear quadrilateral elements; the interface is tracked on this mesh using marker points that define the interface position at each time. The markers are placed to define the initial interface position to a prescribed tolerance; as the interface develops, marker points are added or deleted so as to maintain the specified resolution. This part of the algorithm has been shown to be second order accurate and is described in Ref.(20). The solute conservation equation is solved on a changing mesh of quadratic triangular elements in the liquid only. A new mesh is generated at each time step that follows the evolving interface; the values from the

previous mesh are interpolated to the new one and a new solute concentration field is solved. The energy and solutal concentration equations are coupled at the interface through Eq. (6), and the solution iterated to convergence using the relative change in interface velocity as the convergence criterion.

3. Interface Numerical Stability

It was mentioned in the introduction that in previous work⁽⁶⁾⁻⁽¹⁶⁾ simplifying assumptions have been made to make the problem more amenable to numerical calculations. The most important of these approximations are the assumption of a known temperature field, and the consideration of very dilute mixtures; both of which directly improve the numerical stability of the models. The assumption of isothermal solidification^{(6),(8)} implies that the solute alone controls the dendritic growth and approximates the case of alloys with large concentrations of solute. Models that utilize a prescribed temperature field that is not modified by the shape of the interface^{(9),(11)} require that the thermal conductivities of the solid and liquid be similar, and that the latent heat released upon solidification be small.

Theoretically, the linear stability analysis of Mullins and Sekerka⁽²³⁾ shows the increased instability of systems with large concentrations of solute. Figure 1(a) shows perturbations growth rate as a function of wavelength for Pb-Sb alloys with initial concentrations of 0.02, 0.2 and 2.2 wt% Sb solidifying at 0.028 mm/s. As the concentration of Sb increases, the most unstable wavelength decreases from approximately 0.08 mm to 0.005 mm; at the same time the growth rate of the most unstable wavelength increases from approximately 0.5 s⁻¹ to 22.0 s⁻¹. Therefore, a calculation at a concentration of 2.2 wt% Sb requires a mesh 16 times finer than that for a concentration of 0.02 wt% Sb to capture the most unstable wavelength, and a time step 44 times smaller to preserve the numerical stability. Figure 1(b) shows the critical and most unstable wavelengths as a function of the initial solute concentration.

Numerically, in the algorithm under consideration, the interface position and velocity are calculated from Eq. (2.b) using quadratic approximations to the temperature gradients in the direction normal to the interface, given by

$$\frac{\partial T_s}{\partial n} \approx \left(\frac{2x-3a}{2a^2} \right) T_1 + \left(\frac{2a-2x}{a^2} \right) T_2 + \left(\frac{2x-a}{2a^2} \right) T_l \quad (7.a)$$

$$\frac{\partial T_l}{\partial n} \approx \left(\frac{2x-3a}{2a^2} \right) T_l + \left(\frac{2x-2a}{a^2} \right) T_3 + \left(\frac{2x-a}{2a^2} \right) T_4 \quad (7.b)$$

(illustrated in one dimension in Fig. 2) where a is a

Fig. 1 Linear stability of the interface for Pb-Sb alloys: (a) perturbation growth rate vs. wavelength; (b) critical (λ_c) and most unstable (λ_m) wavelength vs. initial concentration. The interface velocity is $V = 28 \mu\text{m/s}$.

Fig. 2 One-dimensional schematic and notation for the calculation of the temperature gradients at the interface

user defined distance, x_k are coordinates of the nodes, x_I is the interface position and Eqs. (7.a) and (7.b) are written, respectively, with respect to local coordinate systems located at $x = p_1$ and $x = x_I$ in Fig. 2. In these equations, the interface temperature T_I is needed to evaluate the temperature gradients. The stability of the algorithm depends directly on the strength of the

Table 1 Physical properties of a Pb-Sb alloy

Property	Symbol, Unit	Value
Bulk concentration of Sb	C_0 , wt%	2.2
Solute diffusivity	D_L , m^2/s	1.13×10^{-9}
Solute partition ratio	k	0.312
Eutectic concentration	C_E , wt%	11.2
Slope of liquidus line	m , K/wt%	-6.829
Melting temperature of pure solvent	T_{m0} , K	600.0
Eutectic temperature	T_E , K	523.5
Density	ρ , kg/m^3	1.0416×10^4
Specific heat of solid	c_{PS} , J/kg·K	142.0
Specific heat of liquid	c_{PL} , J/kg·K	151.0
Heat conductivity in the solid	κ_S , J/s·m·K	30.0
Heat conductivity in the liquid	κ_L , J/s·m·K	16.0
Latent heat of fusion	L , J/kg	29775
Surface energy	γ_0 , J/m ²	7.0×10^{-2}
Kinetic mobility	ν , m/s·K	6.67×10^{-3}

constraint on the interface temperature, the best case being if T_i is fixed and constant. In the case of a pure substance, Eq.(5) takes a simpler form. Because T_m becomes constant, it no longer depends on the local solute concentration at the interface through Eq.(6). In this case, the main contribution to change in the interface temperature comes from the surface energy term; the effect of the kinetic mobility and the difference in specific heat can be shown to be small, and even though the interface temperature varies locally, the changes are not significant enough to affect the stability of the calculations. A very different situation arises in the case of a binary alloy, when T_m is given by Eq.(6). Table 1 shows the physical properties of the Pb-2.2 wt% Sb alloy considered in this article. Notice that $m = -6.829$ K/wt% and the equilibrium partition ratio is $k = 0.312$, so at steady-state the interface is expected to reach concentrations of around 7%; this produces a change of up to 48 K in the interface temperature. The strong variability in the interface temperature affects the convergence of the non-linear iteration and makes the calculations considerably more difficult. As a result the time step for the calculation must be made much smaller, and more non-linear iterations are needed for convergence producing a very large increase in the computational time needed to perform the simulations. The above conclusions are illustrated through numerical calculations in the next section. The strong dependence of the computational stability and cost on the initial solute concentration is shown for the Pb-Sb alloys considered in this article.

4. Examples

In the calculations presented below, the surface tension was assumed to exhibit four-fold anisotropy and given by

Table 2 Physical properties of succinonitrile used in the numerical simulations

Property	Symbol, Unit	Value
Specific heat of the solid	ρc_{PS} , J/m ³ ·K	1.97×10^6
Specific heat of the liquid	ρc_{PL} , J/m ³ ·K	1.92×10^6
Heat conductivity in the solid	κ_S , J/s·m·K	0.224
Heat conductivity in the liquid	κ_L , J/s·m·K	0.223
Thermal diffusivity in the solid	α_S , m ² /s	1.15×10^{-7}
Thermal diffusivity in the liquid	α_L , m ² /s	1.16×10^{-7}
Melting temperature	T_{m0} , K	331.25
Latent heat of fusion	ρL , J/m ³	4.54×10^7
Surface energy	γ_0 , J/m ²	8.95×10^{-3}
Kinetic mobility	ν , m/s·K	0.2

$$\gamma = \gamma_0 \left[1.0 + 0.4 \left(\frac{8}{3} \sin^4(2\theta) - 1.0 \right) \right] \quad (8)$$

(see e.g. Refs.(17), (18)), the kinetic mobility is constant. The simulations are done in a rectangular domain starting from a vertical planar front located very close to the left end of the domain; the horizontal boundaries are adiabatic. First we consider solidification of a pure substance using the properties of succinonitrile given in Table 2. The solid is at the equilibrium melting point (331.25 K) and on the left boundary the temperature is fixed at the melting point. The liquid is undercooled by 13.01 K, which yields a Stefan number $St = c_{PL} \Delta T / L = -0.55$, where $\Delta T = -13.01$ K. The right side is held at $T_m + \Delta T$.

The linear stability analysis⁽²¹⁾ predicts the most unstable wavelength λ_m to be approximately proportional to $1/\sqrt{V}$, where V is the initial steady-state velocity of a planar interface. Here we choose the same length scale $\lambda_0 = 27.4 \mu\text{m}$, corresponding to $V = 0.1$ mm/s, that was used in Ref.(22) for similar calculations. The domain is set at $12\lambda_0$ by $8\lambda_0$, and four equally spaced perturbations are introduced by displacing the markers at those locations by $\epsilon = 0.005\lambda_0$. The results (Fig. 3) obtained on a mesh of 600 by 400 bilinear elements show perturbations that quickly evolve into four primary dendrites growing almost parallel to each other and at a nearly steady-state velocity. Behind the dendrite tips coarsening effects can be clearly seen: larger secondary arms grow even larger and smaller ones gradually disappear, resulting in a dendritic structure less detailed than near the tips. The CPU cost of all calculations presented here is summarized in Table 3. This case (Case 1), which is the one with the best interface stability properties, is also computationally the most affordable. The calculations were performed in a 400 MHz SGI Origin 2000 using a single processor at The University of Arizona Computer Center.

Table 3 Summary of computed cases

Case	Material	C_0 (wt%)	Domain (mm)	Temperature Mesh	Concentration Mesh (# of quadratic triangular elements)	No. of Time Steps	Simulation Time(s)	CPU Time (hrs)
1	Succinonitrile	0	0.3288×0.2192	600×400	N/A	25,000	0.0047	21
2	Pb-Sb alloy	0.02	2×0.6	100×30	7,000-15,000	22,000	55	51
3	Pb-Sb alloy	0.2	0.8×0.24	100×30	10,000-20,000	20,000	4	44
4	Pb-Sb alloy	2.2	0.8×0.24	100×30	10,000-25,000	130,000	26	370

Fig. 3 Solidification of succinonitrile from a planar front perturbed at four discrete locations. From left to right, interface location at times 0.0016, 0.0032 and 0.0047 s. The spatial dimension unit is μm .

The next calculations are for Pb-Sb alloys using the properties given in Table 1 and illustrate the increasing computational difficulty as the alloys go from being very dilute to concentrations of several weight percent. Three initial concentrations (C_0) are considered here, 0.02, 0.2 and 2.2 wt% Sb; these make up Cases 2, 3 and 4 respectively in Table 3.

In all cases of the Pb-Sb alloys the melt is initially in a linear temperature gradient. A cooling rate is imposed on the left boundary, and the initial temperature gradient is applied as boundary condition on the right boundary. A one-dimensional model was run first. The two-dimensional calculations used the one-dimensional results; i.e., the temperature and concentration fields were used as initial conditions, keeping the same boundary conditions along the vertical boundaries. In Cases 2 and 3, the one-dimensional calculation was carried out until a steady-state was reached; in Case 4 the one-dimensional calculation was stopped before it attained steady-state. For Case 2, the same material properties as used in Ungar and Brown⁽⁹⁾ are chosen for comparison with their results. The temperature gradient is 1.008 K/mm and the cooling rate is 0.039 K/s . The latter were chosen so that with the same reference length $\lambda_0 = 0.1 \text{ mm}$, the dimensionless temperature gradient G and the growth rate as expressed by the Péclet number $Pe = V\lambda_0/D_L$, are the same as in Ref. (9), these are $G = 1.68 \times 10^{-4}$ and $Pe = 1.4$. The steady-state temperature and

Fig. 4 Solidification of Pb-0.02 wt% Sb from a planar front perturbed with a cosine wave. Interface position is shown at intervals of 10 s up to 40 s. The spatial dimension unit is mm.

solute concentration fields obtained from a one-dimensional calculation were then used in two-dimensions as initial conditions on a planar front, which is then perturbed with a cosine perturbation of wavelength λ_0 and amplitude $0.01\lambda_0$. A mesh of 100 by 30 elements was used for the temperature field in a domain of size $20\lambda_0$ by $6\lambda_0$. The solute concentration was solved in a variable mesh of quadratic triangular elements with sides less than 20% of the solutal diffusion length $l = \lambda_0/Pe$ at the interface, the initial mesh contained close to 12 000 elements. Results of the calculation are shown in Fig. 4 for up to 40 s of solidification. The cellular structure develops with wavelength $\lambda_0 = 0.1 \text{ mm}$, which is very near to the most unstable wavelength predicted by linear stability theory. The interface position is plotted every 10 s. As the interface develops the grooves between cells become deeper and droplets form at the bottom of the grooves. Comparing Fig. 4 with Fig. 10 in Ref. (9), it can be observed that they are in good qualitative agreement, but some differences can be observed. In the present results the top of the cells are flatter and the grooves are narrower than in Ref. (9). The depth of the cells remains comparable. A quantitative comparison is not feasible, because the model in Ref. (9) assumed a prescribed temperature field and therefore neglected the latent heat of solidification. In the present model the latent heat is an essential part of the mechanism that drives the interface. The inclusion of the latent heat also changes the linear stability characteristics of the interface, e.g., at $Pe = 0.8$ (used in Ref. (9)) the linear stability analysis predicts a stable system if latent heat is included, and an unstable system if it is not. A second difference involves the inclusion in Ref. (9) of

Fig. 5 Solidification of Pb-0.2 wt% Sb from a randomly perturbed planar front; (a) interface position at intervals of 0.2 s; (b) solute concentration at 4 s (the white line is the interface); (c) temperature field at 4 s

solutal diffusion in the solid, which has been neglected in this model.

Case 3 is calculated in the same way as Case 2, except that the initial solute concentration is now 0.2 wt% Sb, and the domain is smaller than in Case 2 reflecting the smaller preferred unstable wavelength. The interface position at intervals of 0.2 s, up to 4 s is shown in Fig. 5(a). A dendritic structure develops after the initial small protuberances are replaced very rapidly by an also short-lived cellular structure. In Fig. 5(b) the solute concentration field at 4 s is shown, the solid-liquid interface is shown in white. In the solid the concentration is calculated from the interface concentration as $C_s = kC_L$; the buildup of solute concentration in the mushy zone can be observed as well as some microsegregation in the solid. Figure 5(c) shows the temperature field at 4 s. The effect of the latent heat can be clearly observed as a difference in the temperature gradient between the liquid and the mushy zone, and at the tips of the dendrites the temperature field is highly distorted and gradients in the vertical direction are also present. This clearly indi-

Fig. 6 Solidification of Pb-2.2 wt% Sb from a planar interface perturbed by a cosine wave. Interface position at intervals of 1 s up to 26 s

cates that assuming that the temperature field exhibits a linear gradient in the x -direction is not a valid approximation for solutions that release significant amounts of latent heat.

In Case 4 the temperature gradient is 10 K/mm, which is more in line with those used in the actual casting of turbine blades. The cooling rate is 0.1 K/s, and the computational domain is 0.8 mm by 0.24 mm. The one-dimensional simulation was carried until $C_L = 4.69$ wt% Sb and $V = 3.79 \mu\text{m/s}$. A mesh of 100 by 30 bilinear elements was used to calculate the temperature; the mesh for the solute concentration initially contained 80 quadratic triangles at the interface. The total number of triangular elements in a mesh varied between 10 000 and 25 000 during the simulation. The results for the interface location are shown in Fig. 6 up to 26 s, at time intervals of 1 s. As shown in Table 3, this simulation is by far the most expensive, because very small time steps have to be used. We are also restricted to a very small computational region that does not allow the solidifying morphologies to reach the point where a well-defined dendrite has been produced. We see first a large number of small protuberances appear on the interface; these protuberances quickly develop into a cellular structure that in turn changes to the dendritic morphologies observed in Fig. 6. Finally the tips accelerate enough to form well-defined dendrites.

5. Conclusions

Simulations of dendritic solidification of binary alloys have been examined based on the computational difficulty introduced by the presence of high concentrations of solute in the mix, that affects the stability of the solid-liquid interface. The numerical model described in Refs. (17) and (20) has been used to perform two-dimensional simulations of solidification of Pb-Sb alloys, for solutions with concentrations varying over two orders of magnitude, to illustrate the increasing cost of the simulations as the solute concentration increases to the level of a few wt% Sb (see Table 3). Even though some of these simulations are expensive, it has been shown that they are within the capabilities of current numerical

models and as we learn how to compute in this environment full three-dimensional simulations are not far ahead. Presently the algorithm is being extended to include the effect of variable density and convection.

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