A Phase-Field Approach for Liquid to Solid Phase Transformation for Binary Ni-Cu Alloy

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1. Phase-Field Approach and Solidification

When modeling phase transitions within a material, it is necessary to solve a set of equations governing the thermodynamics of the problem at hand. The equations involved are meant to describe diffusion in the bulk phases and the intricate conditions at the moving phase boundary. As an example, when dealing with solidification in a pure material, the governing set of equations is called the "Stefan problem", given by

$$\dot{T} = \alpha \nabla T \qquad\qquad x \in \Omega_{L,S}(t)$$

$$H_{fus}\, \boldsymbol{v}_n = -c\alpha \left(\frac{\partial T}{\partial \boldsymbol{n}}\right)^{solid}_{liquid} \qquad\qquad x \in \Gamma(t)$$

$$T = T_m - \frac{\sigma T_m}{H_{fus}}\,\kappa - \frac{v_n}{\mu} \qquad\qquad x \in \Gamma(t)$$

where $\Omega_{L,S}$ is the bulk region of the solid/liquid phase and Γ is the boundary separating. Further, $T = T(\boldsymbol{x}, t)$ is the temperature field of the system with T_m its melting temperature, c its heat capacity, H_{fus} the latent heat of fusion, α the thermal diffusivity, v_n the normal velocity of the interface, κ the radius of curvature of the interface, σ the surface tension and μ the inter facial mobility. The first equation governs the heat transfer in the two phases, the second equation, which describes the condition of heat balance at the interface, states that the normal velocity of the interface is proportional to the discontinuity in the temperature gradient. The last equation, called the Gibbs-Thomson relation, captures the change of melting temperature at the interface due to kinetic and capillary effects, i.e. interaction between solid and liquid phase boundaries. In the equations defining the Stefan problem, the width of the interface is treated as a mathematical surface; meaning that the width of the interface is non-present. This is referred to as a "sharp interface." It is generally assumed, when modeling solidification, that the interface between two phases is in local thermodynamically equilibrium. Moreover, a pure melt can exist at a temperature $T < T_m$ but will begin to solidify if a disturbance to the system in the shape of a crystal seed is introduced. There are two competing driving forces that govern the growth of the seed; what they have in common is that they strive to reduce the overall free energy of the system to a minimum. The first one is the release of latent heat from the growing crystal where the faster the latent heat is conducted away, the faster the interface can evolve. It is thus desirable for the interface to evolve so as to maximize its surface area, but the interface also comes with a surface energy which in turn implies that the interface would evolve so as to minimize its surface area. The growth of the interface is thus governed by a compromise between these two phenomena. A different situation arises when dealing with the solidification of an alloy; here one has to consider another type of diffusion apart from the thermal diffusion: namely the diffusion of chemical species.

However, the rate of thermal diffusion in a metal is much higher than that of the solute diffusion; it is thus the latter of the two effects which is dominant in governing the growth of the interface. The rejection of solute from the interface is analog to the release of latent heat in the thermal case. Additionally, the fact that the thermal diffusion operates on a timescale much smaller than the chemical diffusion makes the numerics involved in simulation of non-isothermal solidification nontrivial.

The phase-field method, which was introduced by Cahn and Hilliard [1] to model phase separation in a binary mixture of fluids, introduces a parameter $\phi(x, t)$ defined on a continuum field; it does not have any discontinuities but takes on a fixed value in the bulk phases and varies continuously across the interface which is of finite width. The values of the parameter - are used to describe the order of the phases where a disordered phase, i.e. liquid would be defined by $\phi = a$ and an ordered phase, i.e. solid would be defined by $\phi = b$. The parameter ϕ is therefore commonly referred as the order parameter. The phase field model for solidification consists of an equation for the evolution of the order parameter coupled with equations governing the dynamics of diffusion; in these equations, the thermodynamic and kinetic coefficients are tuned so that the phase-field model becomes equivalent to the original sharp interface model in the limit when the interface width $w_\phi \rightarrow 0$. The main advantage of the phase-field model is that the explicit tracking of the interface is no longer required.

3. Non-isothermal solidification of a Ni-Cu alloy

In this section, a model which includes thermal effects is used to simulate the dendrite growth of a crystal seed in an undercooled melt of a binary Ni-Cu alloy. Adding an equation for the heat transfer does not only produce a more realistic model of the solidification process but also allows one to study the effects of boundary heat flux, which is of great importance to, for example, metal casting in the industries. The model is given in [2] and not repeated here. The phase-field and diffusion equation were discretized using the finite volume method and an explicit Euler scheme. When it comes to solving the heat equation, it becomes problematic due to the vast difference in the diffusivity rate of the solute and temperature field. The temperature field evolves much more rapid than the solute field and to keep numerical stability, with explicit schemes, the time step must be chosen several orders of magnitude smaller compared to when solving the isothermal model which, in turn, leads to a dramatic increase in simulation time. We overcome this problem by the semi-implicit ADI method which, in the two-dimensional case, alternates between treating grid point values implicit in the x-direction and explicit in the y-direction and vice versa, hence the name alternating direction implicit. A typical simulation starts from a undercooled, supersaturated melt with an initial fraction of $c = 0.4083$ Ni and an initial temperature $T = 1574\ K$. A small crystal seed was placed in the center of the simulation grid. The growth of the dentrite for two different thermal boundary conditions at $t = 2.6\ ms$ are depicted in Fig. 1.
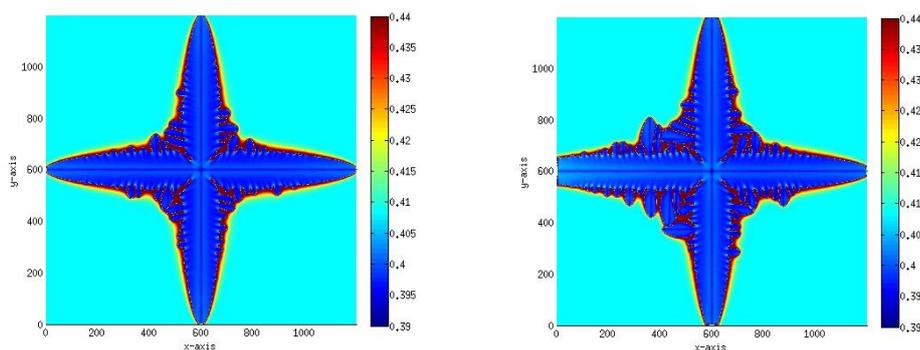
Figure 1. Growth of a dentrite in a Ni-Cu alloy. Left: adiabatic boundary conditions. Right: a negative heat flux at left boundary is applied, thus the dentrite growth faster on the left hand side.

6. References

[1] J. W. Cahn and J. E. Hilliard (1958). Free energy of a nonuniform system I. *J. Chem. Phys.*, **28**:258.
[2] L. Du and R. Zhang (2014). Phase field simulation of dendrite growth with boundary heat flux. *Integrating Materials and Manufacturing Innovation*, **3**:18.