Bachelor of Science (Research)
Material Science

Final Year Project Report

To understand the influence of driven solutal convection on dendrite growth.

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Acknowledgements

Synopsis

Solidification is one of the major processing routes among the various materials processing techniques, where in there is phase transformation from a liquid to one or more solid-phases. The process gives rise to a variety of microstructures, arising out of dendritic, eutectic, peritectic and monotectic reactions. It is well known that properties of a material can be linked to its structure at the microscopic scale. Thus, in order to manufacture a cast product with desired properties, it is essential to determine the influence of the processing parameters on microstructural evolution. Here, modelling of complex microstructural evolution allows for the determination of the useful process $\rightarrow$ structure and parameter $\rightarrow$ structure correlations.

Being a first order phase transition, the solidification reaction involves a transfer of heat/mass across the interface between the solid and the liquid phases concomitant with diffusion in the bulk liquid and the solid phases. Any modeling technique describing this phenomenon will require, that in addition to the global boundary conditions controlling the heat/mass exchange, it self-consistently is able to integrate the transport processes at the moving interface between the evolving phases with those in the bulk.

Classically, such problems were treated with *Sharp-interface* methods, wherein, separate transport equations are solved in the respective bulk phases, while the appropriate (*Stefan-boundary*) conditions are imposed at the interface nodes which therefore need to be marked after each time iteration. The method becomes cumbersome in case of complex morphological evolution which is commonly encountered in most solidification reactions, where explicit interface tracking becomes computationally expensive and resolving sharp curvatures necessitates the use of finer meshes.

In this context, we resort to the phase field method - a state-of-the art technique, developed in the past three decades which obviates the need for tracking of the moving interfaces. Succinctly, the method describes evolution equations for order parameters varying smoothly between the various phases, thereby representing phase evolution. The various boundary conditions at the moving interface are self-consistently described in the transport equations describing solutal/heat/momentum transport, which are defined globally. The transport equations are coupled with the evolution equations for the order parameter. This class of methods has been applied to a variety of phase transformation reactions.

In this present study, we investigate the influence of convection on dendritic

growth. Microstructural evolution during solidification is strongly influenced by flow in the liquid phase. The effect is due to large scale transport of mass or heat by the fluid currents. The inclusion of convection leads to the modification of the transport equations.

In the phase field method the system properties are expressed using a set of phasefield varibales that vary continuously within a bound across all the phases in the system. The thrmodynamics and transport properties of the system are coupled with or expressed in terms of the dynamics of these phasefield variables.

The time evolution of the phase field variables is given by a set of coupled partial differential equations, one equation for each variable. The equation are derived according to the principle of non-equilibrium thermodynamics.They are so chosen that the free energy of the system decreases with time and mass is conserved for all components. Likewise transport properties within the bulk as well as the interface are incorporated to be consistent with the dynamics of these phasefield variables. Numerical solution of these PDE's yields the temporal evolution of the phasefield variables, which is a representation of the morphological evolution microstructutre in the system.

In order to develop a model for soldification, we make use of an order parameter, ϕ, as the phase field variable. ϕ is unity in the solid phase, zero in the liquid phase and takes intermediate values at the interface. A free energy functional for the sytem is described its variational derivative gives the evolution equation for ϕ and other state variables. Apart from this, constructs in terms of ϕ are used to describe system properties like anisotropy.

In order to model fluid flow we need to set up Pressure and Velocity fields which are coupled with the phasefield variable. The evolution of the appropriately coupled velocity fields is computed through direct numerical simulations(DNS) of the navier stokes equation. The pressure field at each time step to ensure continuity in the velocity fields.

In the subsequent chapters we have described in detail how each element of the phase field mdoel was set up. We have further described how the equations were descretised and what numerical computation techniques were used. Finally, we have discussed results from our simulations and laid out analysis and plan for further action.

Literature survey and importance of dendritic solidification

The phase field modelling has been used for alloy solidification and for the simulation of micorstructure for more than a decade.

A Phase field model for solidification

The application of the phase field method starts with the creation of a functional which includes the material properties involving both the surface properties of the interfaces in the system and the thermodynamic energy of the bulk phase in the system. A variational derivative of this functional with respect to any of the changing phasefield variables, gives us the driving force for the change. Depending on whether the we are treating a pure or a multi component system this driving force is a function of just the temperature or includes the composition of the various components as well.

3.1 Modelling alloy Solidification

In this section, we we will see how to model the problem of solidification based on a reaction diffusion type model where in the reaction is modelled using an Allen-Cahn type equation while the diffusion term is modelled using a Cahn-Hilliard type model

We shall first consider the case of solidification of binary isomorphous alloy, wherein at a given undercooling, a driving force due to deviation of the diffusion potential from the equilibrium, results in solute rejection at the interface and expansion of the solidification front.

So, in principle we are going to need two evolution equations, one for the field: order-parameter ϕ whose evolution will indicate phase transformation from liquid ($\phi = 0$) to $\phi = 1$ that is the solid; the second, will be the evolution of the diffusion potential field μ. Lets see how we go about putting the things together.
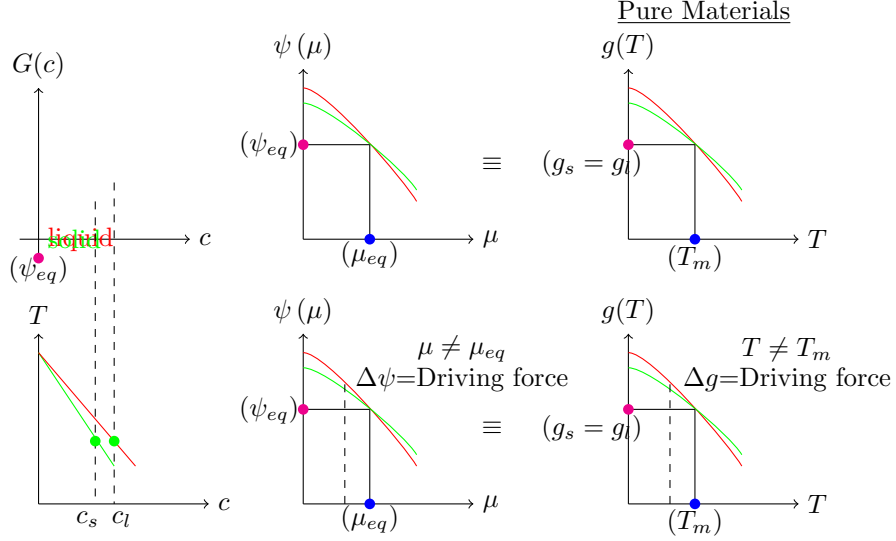
Assuming conditions of local thermodynamic equilibrium(LTE), the driving force for phase transformation in alloys is the difference of the grand-potentials of the solid and the liquid phases (or the difference of the chemical potentials of any of the chemical entitities depending on whether we start from a Helmholtz free-energy density or a Gibbs-free energy density in the functional).

In the present description, we adopt a Helmholtz free energy density which then results in the driving force for solidification being described as $\Psi_l - \Psi_s$ This is analogous to the case of pure material solidification where the driving force is given by $g_l - g_s$. However, while the state-variable in the case of pure material solidification is the temperature T, it becomes the diffusion-potential μ in the case of binary alloy solidification.

This analogy can be well appreciated from the diagramm in Fig 3.1.

At equilibrium, the common-tangent construction gives the equilibrium compositions of the solid and liquid, which can be read from the equilibrium phase diagram. This same equilibrium can also be represented as the intersection of the grand-potential densitities $\Psi(\mu)$ expressed as a function of the diffusion-potentials μ, where $\Psi_s(\mu_{eq}) = \Psi_l(\mu_{eq})$, in the same manner as $g_s(T_m) = g_l(T_m)$ for the case of pure materials. Away from equilibrium $\mu \neq \mu_{eq}, \Psi_l \neq \Psi_s$,and we have a driving force for phase transition given by the difference of the grand-potential densities.

Therefore, using this above analogy we can write down the driving force for phase transition for alloys purely in terms of the diffusion potential μ by performing a Taylor series expansion around equilibrium, until first order as follows,

$$\Psi_s(\mu) = \Psi_s(\mu_{eq}) + \frac{\partial \Psi_s}{\partial \mu}(\mu - \mu_{eq}) \tag{3.1}$$

$$\Psi_l(\mu) = \Psi_l(\mu_{eq}) + \frac{\partial \Psi_l}{\partial \mu}(\mu - \mu_{eq}). \tag{3.2}$$

From the fig. 3.1 it is clear that we can express the grand-potential as

$\Psi(\mu) = g(c(\mu)) - \mu c(\mu)$, which is the essentially the *Legendre transform* of g. Therefore, we can derive,

$$\frac{\partial \Psi}{\partial \mu} = \frac{\partial g}{\partial c}\frac{\partial c}{\partial \mu} - \mu\frac{\partial c}{\partial \mu} - c \qquad (3.3)$$

$$= -c, \qquad (3.4)$$

where we have used $\frac{\partial g}{\partial c} = \mu$. Clearly, then we can write the driving force for solidification $\Psi_l - \Psi_s$ as,

$$\Psi_l - \Psi_s = (c_s^{eq} - c_l^{eq})(\mu - \mu_{eq}), \qquad (3.5)$$

where we have used $\Psi_l(\mu_{eq}) = \Psi_s\mu_{eq}$ and $\frac{\partial \Psi_l}{\partial \mu}\bigg|_{\mu_{eq}} = -c_l^{eq}$ and $\frac{\partial \Psi_s}{\partial \mu}\bigg|_{\mu_{eq}} = -c_s^{eq}$. This relation can be transmitted in the evolution equation for the phase-field variable ϕ as,

$$\frac{\partial \phi}{\partial t} = -M\left(\frac{\partial f_o}{\partial \phi} - 2\kappa\nabla^2\phi\right) - M\underbrace{(c_l^{eq} - c_s^{eq})(\mu - \mu_{eq})\frac{\partial h(\phi)}{\partial \phi}}_{driving\,force}. \qquad (3.6)$$

The evolution equation of the coupled variable μ can be derived using mass conservation. We have chosen interpolating polynomial $h(\phi)$ such that:

$$h(\phi) = \phi^2(3 - 2\phi) \qquad (3.7)$$

$$\frac{\partial h(\phi)}{\partial \phi} = 6\phi(1 - \phi) \qquad (3.8)$$

The local composition can be written as,

$$c = c_s h(\phi) + c_l(1 - h(\phi)), \qquad (3.9)$$

More rigourously, the above relation derives from the interpolation of the grand-potential densities $\Psi(\mu) = \Psi_s(\mu)h(\phi) + \Psi_l(\mu)(1 - h(\phi))$, and taking the derivative with respect to the diffusion potential leads us to the relation, $\frac{\partial \Psi}{\partial \mu} = \frac{\partial \Psi_s}{\partial \mu}h(\phi) + \frac{\partial \Psi_l}{\partial \mu}(1 - h(\phi))$.

Substituting the above relation between the compsition and the diffusion-potential into the mass-conservation equation we have,

$$\frac{\partial c}{\partial t} = \nabla \cdot (M\nabla\mu) \qquad (3.10)$$

$$\left(\frac{\partial c_s}{\partial \mu}h(\phi) + \frac{\partial c_l}{\partial \mu}(1 - h(\phi))\right)\frac{\partial \mu}{\partial t} = \nabla \cdot (M\nabla\mu) - (c_s(\mu) - c_l(\mu))\frac{\partial h(\phi)}{\partial t}. \qquad (3.11)$$

Here, the first term on the right hand side represents the diffusive flux, while the second term is the source term for phase transformation. It corresponds to the amount of mass transfer occuring as a result of solute distribution during phase transformation.

Thus, we are able to derive the evolution equations for phase transformation of pure materials and alloys with a similar starting point of first deriving the driving forces in terms of the relevant state variables and then coupling it with the appropriate conservation equations.

Equations

$\phi = 1 \Longrightarrow$ solid $\phi = 0 \Longrightarrow$ liquid

$$h(\phi) = \phi^2 \left(3 - 2\phi\right) \tag{4.1}$$

$$\frac{\partial h(\phi)}{\partial \phi} = 6\phi \left(1 - \phi\right) \tag{4.2}$$

$$\frac{\partial \phi}{\partial t} = -M \left(\frac{\partial f_o}{\partial \phi} - 2\kappa \nabla^2 \phi \right) - \underbrace{M \left(c_l^{eq} - c_s^{eq}\right) \left(\mu - \mu_{eq}\right) \frac{\partial h\left(\phi\right)}{\partial \phi}}_{driving force}. \tag{4.3}$$

$$\frac{\partial c}{\partial t} = \nabla \cdot \left(M \nabla \mu\right) \tag{4.4}$$

$$\left(\frac{\partial c_s}{\partial \mu} h\left(\phi\right) + \frac{\partial c_l}{\partial \mu} \left(1 - h\left(\phi\right)\right) \right) \frac{\partial \mu}{\partial t} = \nabla \cdot \left(M \nabla \mu\right) - \left(c_s\left(\mu\right) - c_l\left(\mu\right)\right) \frac{\partial h\left(\phi\right)}{\partial t}. \tag{4.5}$$

$$c^l\left(\mu\right) = \mu \tag{4.6}$$

$$c^s\left(\mu\right) = kc^l = k\mu \tag{4.7}$$

$$\Psi_l - \Psi_s = \left(c_s^{eq} - c_l^{eq}\right) \left(\mu - \mu_{eq}\right) \Rightarrow \Psi_l - \Psi_s = (k-1)\mu \left(\mu - \mu_{eq}\right) \tag{4.8}$$

$$= \left(\frac{\partial c_s}{\partial \mu} h\left(\phi\right) + \frac{\partial c_l}{\partial \mu} \left(1 - h\left(\phi\right)\right) \right) \tag{4.9}$$

$$\left(\frac{\partial \mu}{\partial t} = \nabla \cdot \left(M \nabla \mu\right) - \left(c_s\left(\mu\right) - c_l\left(\mu\right)\right) \frac{\partial h\left(\phi\right)}{\partial t} \tag{4.10}$$

$$\frac{dc}{dt} = \frac{\partial c}{\partial t} + \frac{\partial c}{\partial x}\frac{\partial x}{\partial t} + \frac{\partial c}{\partial y}\frac{\partial y}{\partial t} + \frac{\partial c}{\partial z}\frac{\partial z}{\partial t} = \frac{\partial c}{\partial t} + \vec{v} \cdot \nabla c = \nabla \cdot \left(M \nabla \mu\right) \tag{4.11}$$

4.1 Fluid Flow

The navier stokes equation for continuity and momentum transport

$$\frac{\partial \vec{u}}{\partial t} = -\nabla P + \nabla^2 \left(\vec{u}^2 \right) - \vec{u} \cdot \nabla \left(\vec{u} \right) \tag{4.12}$$

Descretisation and Numerical Methods

Results and Discussion

Conclusion

Bibliography