

Phase field analysis of Eutectic Breakdown

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Abstract

In this paper an isotropic multi-phase-field model is extended to include the effects of anisotropy and the spontaneous nucleation of an absent phase. This model is derived and compared against a published single phase model. Results from this model are compared against results from other multi-phase models, additionally this model is used to examine the break down of a regular two dimensional eutectic into a single phase dendritic front.

1 Introduction

Phase field modelling has been used to study both single phase ([1],[2],[3],[4]) systems (single phase models have only one solid phase with the liquid and hence require only a scalar phase variable ϕ) such as the growth of dendrites and multiphase systems ([5],[6],[7],[8],[9]) such as eutectic, peritectic and monotectic solidification. The general concept of all of these models is that rather than a sharp solid-liquid (or solid-solid) boundary, these interfaces between different phases are smooth and continuous. The amount of a particular phase present is given by the phase field variable ϕ . Multiphase systems contain three or more phases and because of this require one phase variable for each phase often written as a vector $\underline{\phi}$ where ϕ_i represents the amount of phase i .

Eutectic microstructures form in alloys when two or more phases grow simultaneously in a co-operative manner. The majority of industrial casting alloys are eutectic or near eutectic in composition. There is a wide variation in the features of eutectic systems, the number of solid phases present limited to the number of alloy components. Recently there has been an increased interest in ternary eutectics for lead-free solder. As well as the number of phases present, the amount of each phase and the entropy of fusion of the phase will significantly effect what structures are formed. With roughly equal amounts of each phase present, lamellar like structures are most prevalent. The theory behind the growth of these was described by Jackson and Hunt [10], while low entropies of fusion will tend to produce regular morphologies. If the amounts of each phase vary significantly then the structures will tend to be more fibrous and higher entropies will tend to make the boundaries between phases more faceted. Eutectics can grow in a similar manner to constrained growth dendrites, without the dendritic arms, in this situation the eutectic equivalent of the primary arm spacing is roughly 1/10 that of the dendritic case. This means that the specific surface area between eutectic phases is significantly greater than for dendritic structures. Additionally, at higher undercooling values the lamellar eutectic

structures have been noted to break down [11] and roughly spherical grains of one phase form within the second phase, furthermore the growth of eutectics and spherical grains within dendrites has been reported [12].

For a given undercooling, ΔT , there is a minimum eutectic spacing for which growth can occur, this is referred to as λ^* , additionally for each value of ΔT there is a maximum eutectic growth velocity (v_0). The maximum velocity can be given in terms of the minimal spacing and the undercooling as follows:

$$\begin{aligned} v_0 &\propto (\Delta T)^2 \\ v_0 &\propto \left(\frac{1}{\lambda^*}\right)^2. \end{aligned}$$

Combining the above gives

$$\lambda^* \propto \frac{1}{\Delta T}, \quad \text{Or,} \quad \lambda^* \Delta T = c, \quad (1)$$

where c is a constant. As the undercooling is increased the minimum interlamellar spacing decreases, thus giving a larger area of interface within the system. This in turn results in a higher total surface energy within the system which will ultimately lead to an unstable system. At this point the coupled growth of eutectics will break down giving rise to single phase growth such as dendrites which grow ahead of the eutectic front.

2 The Model

The model presented here is based upon that derived by Steinbach et. al [8], which this has been extended to include the effects of anisotropic growth. Additionally it is shown that this reduces to the anisotropic single phase model presented by Warren and Boettinger [1]. The following equation gives the free energy functional for this multiphase system.

$$F = \int_V \{G(\underline{\phi}, \underline{\nabla}\phi) + f(\underline{\phi}, c, T)\} dV. \quad (2)$$

The gradient energy term, $G(\underline{\phi}, \underline{\nabla}\phi)$, is given by:

$$G(\underline{\phi}, \underline{\nabla}\phi) = \sum_{j,k(j<k)}^N \left[\frac{\epsilon_{jk}^2 \eta_{jk}^2}{2} |\phi_j \underline{\nabla}\phi_k - \phi_k \underline{\nabla}\phi_j|^2 \right],$$

The following gives the free energy density term, $f(\underline{\phi}, c, T)$, for this system ([1],[6],[7]).

$$\begin{aligned} f(\underline{\phi}, c, T) &= \sum_{j,k(j<k)}^N [(cW_{jk}^B + (1-c)W_{jk}^A)\phi_k^2\phi_j^2] \\ &\quad - \sum_{j=1}^N [(cm_j^B + (1-c)m_j^A)\phi_j^3(6\phi_j^2 - 15\phi_j + 10)] \\ &\quad + \frac{RT}{\nu_m}(c \ln(c) + (1-c)\ln(1-c)). \end{aligned} \quad (3)$$

The vector $\underline{\phi}$ contains the proportion of each phase present, ϕ_1 represents the liquid phase while all higher elements represent solids. The variable c is the concentration variable, both ϕ_i and c vary from $0 \rightarrow 1$. The ϵ_{jk} is a symmetric matrix containing gradient energy coefficients for the j/k interface. η_{jk} is a function of θ_{jk} based upon the orientation of the j/k interface (this term is only needed for anisotropic simulations and in the case of an isotropic system η_{jk} reduces to 1). The angle θ_{jk} gives the orientation of the j/k interface relative to a fixed (but arbitrary) direction. The variable W_{jk} is a symmetric matrix of values which are dependent upon the double-well potential barrier between phases j and k , while the variable m_j relates to the undercooling of phase j . The time evolution of phase and solute is given by

$$\begin{aligned} \frac{\partial \phi_i}{\partial t} &= -M \frac{\delta F}{\delta \phi_i} \\ &= -M \left(\frac{\partial F}{\partial \phi_i} - \nabla \cdot \frac{\partial F}{\partial \nabla \phi_i} \right) \end{aligned} \quad (4)$$

$$\begin{aligned} \frac{\partial c}{\partial t} &= \nabla \cdot \left\{ D(\underline{\phi}) \left[c(1-c) \nabla \left(\frac{\delta F}{\delta c} \right) \right] \right\} \\ &= \nabla \cdot \left\{ D(\underline{\phi}) \left[c(1-c) \nabla \left(\frac{\partial F}{\partial c} \right) \right] \right\}, \end{aligned} \quad (5)$$

where M is a constant based upon interface mobilities and $D(\underline{\phi})$ is a weighted sum of the diffusivities of each phase given by

$$D(\underline{\phi}) = \sum_{i=1}^N [D_i \phi_i].$$

It should be noted that in Equation (5) the variational derivative has been replaced with a partial derivative. The elements of the vector $\underline{\phi}$ must also obey the following condition

$$\sum_{j=1}^N \phi_j = 1. \quad (6)$$

It was shown by Steinbach et. al. [8] that

$$\frac{\partial \phi_j}{\partial \phi_i} = 1 \text{ for } i = j \text{ and } \frac{\partial \phi_j}{\partial \phi_i} = -1 \text{ for one } j \text{ such that } j \neq i. \quad (7)$$

Additionally

$$\frac{\partial \frac{\partial \phi_j}{\partial x}}{\partial \frac{\partial \phi_i}{\partial x}} = 1 \text{ for } i = j \text{ and } \frac{\partial \frac{\partial \phi_j}{\partial x}}{\partial \frac{\partial \phi_i}{\partial x}} = -1 \text{ for one } j \text{ such that } j \neq i. \quad (8)$$

These relations will be required to evaluate the variational differentials in Equations 4 and 5. For ease of reading the following notation will be used [1]

$$\frac{\partial \phi_j}{\partial x} = \phi_{jx}.$$

Equation (4) can be rewritten as

$$\frac{\partial \phi_i}{\partial t} = -M \left(\frac{\partial G}{\partial \phi_i} + \frac{\partial f}{\partial \phi_i} - \frac{\partial}{\partial x} \left(\frac{\partial G}{\partial \phi_{ix}} \right) - \frac{\partial}{\partial y} \left(\frac{\partial G}{\partial \phi_{iy}} \right) \right). \quad (9)$$

Using the assumptions in Equations (7) and (8) it can be shown that

$$\frac{\partial G}{\partial \phi_i} = 0 \quad (10)$$

$$\frac{\partial f}{\partial \phi_i} = \sum_{j \neq i}^N [2W_{ij}\phi_i\phi_j(\phi_j - \phi_i) + 30(m_j - m_i)\phi_i^2\phi_j^2] \quad (11)$$

$$\frac{\partial G}{\partial \phi_{ix}} = - \sum_{j \neq i}^N [\epsilon_{ij}^2(\eta_{ij}^2 r_{ij}^x - \eta_{ij}\eta'_{ij} r_{ij}^y)] \quad (12)$$

$$\frac{\partial G}{\partial \phi_{iy}} = - \sum_{j \neq i}^N [\epsilon_{ij}^2(\eta_{ij}^2 r_{ij}^y + \eta_{ij}\eta'_{ij} r_{ij}^x)], \quad (13)$$

where $W_{ij} = cW_{ij}^B + (1-c)W_{ij}^A$, $m_j = cm_j^B + (1-c)m_j^A$ and $\eta'_{ij} = \frac{\partial \eta_{ij}}{\partial \theta_{ij}}$, where we assume that η_{ij} is only dependant upon θ_{ij} . Additionally, the quantities $\underline{r}_{ij} = \phi_i \underline{\nabla} \phi_j - \phi_j \underline{\nabla} \phi_i = \begin{pmatrix} r_{ij}^x \\ r_{ij}^y \end{pmatrix}$ are defined for convenience.

At this stage the only information assumed about the function η_{ij} is that it is a function of θ_{ij} only, and that the following condition is true

$$\tan(\theta_{ij}) = \frac{r_{ij}^y}{r_{ij}^x}.$$

This allows the anisotropy function to take any form required by the simulation.

The quantities given in Equations (12) and (13) must now be differentiated by x and y respectively in accordance with Equation (9).

$$\begin{aligned} \frac{\partial}{\partial x} \left(\frac{\partial G}{\partial \phi_{ix}} \right) &= - \sum_{j \neq i}^N \left[\epsilon_{ij}^2 \left((2\eta_{ij}\eta'_{ij} r_{ij}^x - \eta_{ij}^2 r_{ij}^y - \eta_{ij}\eta''_{ij} r_{ij}^y) \frac{\partial \theta_{ij}}{\partial x} \right. \right. \\ &\quad \left. \left. + \eta_{ij}^2 \frac{\partial r_{ij}^x}{\partial x} - \eta_{ij}\eta'_{ij} \frac{\partial r_{ij}^y}{\partial x} \right) \right] \\ \frac{\partial}{\partial y} \left(\frac{\partial G}{\partial \phi_{iy}} \right) &= - \sum_{j \neq i}^N \left[\epsilon_{ij}^2 \left((2\eta_{ij}\eta'_{ij} r_{ij}^y + \eta_{ij}^2 r_{ij}^x + \eta_{ij}\eta''_{ij} r_{ij}^x) \frac{\partial \theta_{ij}}{\partial y} \right. \right. \\ &\quad \left. \left. + \eta_{ij}^2 \frac{\partial r_{ij}^y}{\partial y} + \eta_{ij}\eta'_{ij} \frac{\partial r_{ij}^x}{\partial y} \right) \right]. \end{aligned}$$

These two results combine to give the following

$$\begin{aligned} -\underline{\nabla} \cdot \frac{\partial G}{\partial \underline{\nabla} \phi_i} &= \sum_{j \neq i}^N \left[\epsilon_{ij}^2 (2\eta_{ij}\eta'_{ij} (\underline{r}_{ij} \cdot \underline{\nabla} \theta_{ij}) + \eta_{ij}^2 (\underline{\nabla} \cdot \underline{r}_{ij}) \right. \\ &\quad \left. + (\eta_{ij}^2 + \eta_{ij}\eta''_{ij}) \underline{\nabla} \theta_{ij} \cdot \begin{pmatrix} -r_{ij}^y \\ r_{ij}^x \end{pmatrix} + \eta_{ij}\eta'_{ij} \underline{\nabla} \cdot \begin{pmatrix} -r_{ij}^y \\ r_{ij}^x \end{pmatrix} \right) \right]. \quad (14) \end{aligned}$$

All the components of Equation (9) may now be evaluated. The time evolution of the solute is much simpler to find since all the terms can be differentiated

directly, this yields the equation

$$\begin{aligned} \frac{\partial f}{\partial c} &= \frac{RT}{\nu_m} \ln \left(\frac{c}{1-c} \right) + \sum_{j,k(j < k)}^N [(W_{jk}^B - W_{jk}^A) \phi_k^2 \phi_j^2] \\ &\quad - \sum_{j=1}^N [(m_j^B - m_j^A) \phi_j^3 (6\phi_j^2 - 15\phi_j + 10)]. \end{aligned} \quad (15)$$

2.1 Comparison with Single Phase

One of the attractions of this model is the fact that the anisotropic component reduces directly to the single phase equivalent presented by Warren et. al. [1]. In a single phase system rather than a phase vector a single variable ϕ is used, this measures the amount of the liquid phase present (with $1 - \phi$ giving the amount of solid). In order to reduce the multiphase model to a comparable single phase one, it is initially reduced to a set of equations for the solid and liquid phases, ϕ_s and ϕ_l respectively. The final step is to set $\phi_l = \phi$ and $\phi_s = 1 - \phi$ to reduce to the single phase equivalents.

It should be noted that in the single phase case, since there is only one interface, all the interface parameter matrices (eg W_{jk}) are reduced to single variables. Also the term $m_l = 0$ since this is standard for multiphase models ([6],[7]). The first stage of this comparison is to reduce the initial free energy functional given in Equation (2) to a single phase equivalent.

$$\begin{aligned} F &= \int_V \left\{ \frac{\epsilon_{ls}^2 \eta_{ls}^2}{2} |\phi_l \nabla \phi_s - \phi_s \nabla \phi_l|^2 + f(\phi_s, \phi_l, c, T) \right\} dV \\ &= \int_V \left\{ \frac{\epsilon^2 \eta^2}{2} |\phi \nabla (1 - \phi) - (1 - \phi) \nabla \phi|^2 + f(\phi_s, \phi_l, c, T) \right\} dV \\ &= \int_V \left\{ \frac{\epsilon^2 \eta^2}{2} |-\phi \nabla \phi - \nabla \phi + \phi \nabla \phi|^2 + f(\phi_s, \phi_l, c, T) \right\} dV \\ &= \int_V \left\{ \frac{\epsilon^2 \eta^2}{2} |\nabla \phi|^2 + f(\phi_s, \phi_l, c, T) \right\} dV. \end{aligned}$$

This is easily recognizable as a single phase free energy functional [1]

$$\begin{aligned} f(\phi_s, \phi_l, c, T) &= W_{ls} \phi_s^2 \phi_l^2 - m_l \phi_l^3 (6\phi_l^2 - 15\phi_l + 10) \\ &\quad - m_s \phi_s^3 (6\phi_s^2 - 15\phi_s + 10) \\ f(\phi, c, T) &= W(1 - \phi)^2 \phi^2 - m + m\phi^3 (6\phi^2 - 15\phi + 10). \end{aligned}$$

The extra m comes from the fact that $m_l = 0$ and it is the m_s term that remains. The evaluation of $(1 - \phi)^3 (6(1 - \phi)^2 - 15(1 - \phi) + 10)$ gives $1 - \phi^3 (6\phi^2 - 15\phi + 10)$. The decision to set $m_l = 0$ in a multiphase system is an arbitrary decision ([6],[7]), it could have been $m_s = 0$ since the m terms are known relative to each other and a zero point needs to be chosen to write them explicitly. From this it is clear that the 'extra' m in the term above is basically a constant which can take any value depending on where the zero point is taken.

To get from the single phase free energy functional to the evolution equations for phase and solute the following relations are applied [1]:

$$\frac{\partial \phi}{\partial t} = -M \frac{\delta F}{\delta \phi}$$

$$\begin{aligned}
&= -M \left(\frac{\partial f}{\partial \phi} + \frac{\partial G}{\partial \phi} - \nabla \cdot \frac{\partial G}{\partial \nabla \phi} \right) \\
&= -M \left(\frac{\partial f}{\partial \phi} + \frac{\partial G}{\partial \phi} - \frac{\partial}{\partial \phi_x} \frac{\partial G}{\partial \phi_x} - \frac{\partial}{\partial \phi_y} \frac{\partial G}{\partial \phi_y} \right), \tag{16} \\
\frac{\partial c}{\partial t} &= \nabla \cdot \left\{ D(\phi)c(1-c)\nabla \left(\frac{\delta F}{\delta c} \right) \right\} \\
&= \nabla \cdot \left\{ D(\phi)c(1-c)\nabla \left(\frac{\partial f}{\partial c} \right) \right\},
\end{aligned}$$

where $G = \frac{\epsilon^2 \eta^2}{2} |\nabla \phi|^2 = \frac{\epsilon^2 \eta^2}{2} (\phi_x^2 + \phi_y^2)$. Carrying out the above calculations

$$\begin{aligned}
\frac{\partial f}{\partial \phi} &= W2\phi(1-\phi)(1-2\phi) + m30\phi^2(1-\phi)^2 \\
&= 2W\phi(1-\phi)(1-2\phi) + 30m\phi^2(1-\phi)^2 \tag{17}
\end{aligned}$$

$$\frac{\partial G}{\partial \phi} = 0 \tag{18}$$

$$\frac{\partial G}{\partial \phi_x} = \epsilon^2 \eta^2 \phi_x - \epsilon^2 \eta \eta' \phi_y \tag{19}$$

$$\frac{\partial G}{\partial \phi_y} = \epsilon^2 \eta^2 \phi_y + \epsilon^2 \eta \eta' \phi_x. \tag{20}$$

These results can be directly compared with the results in Equations (10)→(13), clearly Equation (10) matches the single phase counterpart given in Equation (18).

Equation (11) represents the derivative with respect to ϕ_i of the free energy density. Equation (17) represents this for a single phase system, to compare the two Equation (11) must first be reduced to a single phase equivalent.

$$\begin{aligned}
\frac{\partial f}{\partial \phi_l} &= 2W_{ls}\phi_l\phi_s(\phi_s - \phi_l) + 30(m_s - m_l)\phi_s^2\phi_l^2 \\
\frac{\partial f}{\partial \phi} &= 2W\phi(1-\phi)((1-\phi) - \phi) + 30(m)(1-\phi)^2\phi^2 \\
\frac{\partial f}{\partial \phi} &= 2W\phi(1-\phi)(1-2\phi) + 30m(1-\phi)^2\phi^2.
\end{aligned}$$

This quite clearly matches Equation (17). Equation (12) also must be reduced to a single phase equivalent before it can be compared with Equation (19).

$$\begin{aligned}
\frac{\partial G}{\partial \phi_{lx}} &= -\epsilon_{ls}^2 (\eta_{ls}^2 r_{ls}^x - \eta_{ls} \eta'_{ls} r_{ls}^y) \\
&= -\epsilon_{ls}^2 (\eta_{ls}^2 (\phi_l \phi_{sx} - \phi_s \phi_{lx}) - \eta_{ls} \eta'_{ls} (\phi_l \phi_{sy} - \phi_s \phi_{ly})) \\
&= -\epsilon^2 (\eta^2 (\phi \frac{\partial(1-\phi)}{\partial x} - (1-\phi)\phi_x) - \eta \eta' (\phi \frac{\partial(1-\phi)}{\partial y} - (1-\phi)\phi_y)) \\
&= -\epsilon^2 (\eta^2 ((-\phi\phi_x - \phi_x + \phi\phi_x) - \eta \eta' (-\phi\phi_y - \phi_y + \phi\phi_y))) \\
&= \epsilon^2 (\eta^2 \phi_x - \eta \eta' \phi_y).
\end{aligned}$$

This matches Equation (19) and it easy to see that the y variation will follow similarly. This is comparable to the intermediate result written by Warren et.

al. [1]. The final stage of the derivation is to find the differentials with respect to x and y given in Equation (16):

$$\begin{aligned}
-\frac{\partial}{\partial \phi_x} \frac{\partial G}{\partial \phi_x} &= -\epsilon^2 (2\eta\eta'\theta_x\phi_x + \eta^2\phi_{xx} - \eta'^2\theta_x\phi_y - \eta\eta''\theta_x\phi_y - \eta\eta'\phi_{xy}) \\
-\frac{\partial}{\partial \phi_y} \frac{\partial G}{\partial \phi_y} &= -\epsilon^2 (2\eta\eta'\theta_y\phi_y + \eta^2\phi_{yy} + \eta'^2\theta_y\phi_x + \eta\eta''\theta_y\phi_x + \eta\eta'\phi_{xy}) \\
-\underline{\nabla} \cdot \frac{\partial G}{\partial \underline{\nabla} \phi} &= -\epsilon^2 \left(2\eta\eta'\underline{\nabla} \theta \cdot \underline{\nabla} \phi + \eta^2 \underline{\nabla}^2 \phi + (\eta'^2 + \eta\eta'') \underline{\nabla} \theta \cdot \begin{pmatrix} -\phi_y \\ \phi_x \end{pmatrix} \right).
\end{aligned} \tag{21}$$

This needs to be compared with a single phase version of Equation (14)

$$\begin{aligned}
-\underline{\nabla} \cdot \frac{\partial G}{\partial \underline{\nabla} \phi_l} &= [\epsilon_{l_s}^2 (2\eta_{l_s}\eta'_{l_s} (r_{l_s} \cdot \underline{\nabla} \theta_{l_s}) + \eta_{l_s}^2 (\underline{\nabla} \cdot r_{l_s}) \\
&\quad + (\eta'_{l_s}{}^2 + \eta_{l_s}\eta''_{l_s}) \underline{\nabla} \theta_{l_s} \cdot \begin{pmatrix} -r_{l_s}^y \\ r_{l_s}^x \end{pmatrix} + \eta_{l_s}\eta'_{l_s} \underline{\nabla} \cdot \begin{pmatrix} -r_{l_s}^y \\ r_{l_s}^x \end{pmatrix})] \\
-\underline{\nabla} \cdot \frac{\partial G}{\partial \underline{\nabla} \phi} &= [\epsilon^2 (2\eta\eta' (-\underline{\nabla} \phi \cdot \underline{\nabla} \theta) + \eta^2 \underline{\nabla} \cdot (-\underline{\nabla} \phi) \\
&\quad + (\eta'^2 + \eta\eta'') \underline{\nabla} \theta \cdot \begin{pmatrix} \phi_y \\ -\phi_x \end{pmatrix} + \eta\eta' \underline{\nabla} \cdot \begin{pmatrix} \phi_y \\ -\phi_x \end{pmatrix})] \\
-\underline{\nabla} \cdot \frac{\partial G}{\partial \underline{\nabla} \phi} &= \epsilon^2 (2\eta\eta' (-\underline{\nabla} \phi \cdot \underline{\nabla} \theta) - \eta^2 \underline{\nabla}^2 \phi \\
&\quad + (\eta'^2 + \eta\eta'') \underline{\nabla} \theta \cdot \begin{pmatrix} \phi_y \\ -\phi_x \end{pmatrix} + \eta\eta' (\phi_{yx} - \phi_{xy})) \\
-\underline{\nabla} \cdot \frac{\partial G}{\partial \underline{\nabla} \phi} &= \epsilon^2 \left(-2\eta\eta' \underline{\nabla} \phi \cdot \underline{\nabla} \theta - \eta^2 \underline{\nabla}^2 \phi - (\eta'^2 + \eta\eta'') \underline{\nabla} \theta \cdot \begin{pmatrix} -\phi_y \\ \phi_x \end{pmatrix} \right).
\end{aligned} \tag{22}$$

This equation is clearly the same as Equation (21), additionally it can be shown that this is the same anisotropic form that was presented by Warren et. al. [1]

3 Noise

Due to the nature of the initialization of the system the random perturbations which are present in real systems are absent from a computational one. If certain features are to be observed then these must be included in the system. One feature which relies on the presence of noise is side arm branching in dendritic systems.

Within the multiphase system there are two types of noise used. The first is a simple interface noise which is analogous to that used extensively in single phase systems and which is required to produce the side arm branching in dendrites. The second type of noise is more complicated; this is a nucleation noise. This is intended to nucleate a small amount of a phase which isn't present, if growth conditions are favorable then this will grow, if they are not then it will shrink. This noise is used to replicate the fact that one solid phase, α , can nucleate a second solid phase β . It is required in some, but not all, of the simulations.

3.1 Interface Noise

The interface noise within this system is handled in a relatively simple manner, analogous to the single phase implementation of Warren et. al. [1]. The following equation describes this.

$$\frac{\partial \phi_i}{\partial t} = -M \left(\frac{\delta F}{\delta \phi_i} + \sum_{j=1}^{i-1} \left[\alpha_{in} r (16 \phi_i^2 \phi_j^2) \frac{\partial f}{\partial \phi_i} \right] \right), \quad (23)$$

where r is a random sample from a Gaussian profile, α_{in} is a constant used to vary the total effect of the noise, a typical value used here is $\alpha = 0.3$ [1]. To maintain the condition set out in Equation (6), when an amount is added to phase i the same amount must be subtracted from phase j .

3.2 Nucleation Noise

The nucleation noise term is added to the system to allow the nucleation of a phase which is not currently present. This addition is made in order to simulate specific microstructures for example the splitting of a series of wide eutectics to form narrower eutectics, examples of this can be seen in the results section as well as in literature [6].

The form of this term is quite similar to the form of the interface noise. It was decided that rather than allow full homogeneous nucleation anywhere within the domain, the nucleation could only occur on a solid-liquid interface. The conditions for the nucleation of phase i can be written as follows.

- Only nucleate phase i if it is not already present, or $\phi_i = 0$.
- Make sure there is an interface between a solid and a liquid phase, this condition can be written as $\phi_l \times \phi_j \neq 0$ and $j \neq l$.

Additionally a thermodynamic multiplier is needed. In the interface noise example it is $\frac{\partial f}{\partial \phi_i}$. Unfortunately because of the form of the ϕ_i multipliers within $\frac{\partial f}{\partial \phi_i}$, this term is zero if $\phi_i = 0$. Therefore the thermodynamic multiplier here uses the undercooling of phase i (m_i) multiplied by the appropriate $\phi_j \phi_l$ interface multiplier. The nucleation noise equivalent of Equation (23) is therefore given by

$$\frac{\partial \phi_i}{\partial t} = -M \left(\frac{\delta F}{\delta \phi_i} + \sum_{j=1}^{i-1} \left[\alpha_{nuc} r (16 \phi_j^2 \phi_l^2) 30 m_i (\phi_j \phi_l) \right] \right), \quad (24)$$

where α_{nuc} is the nucleation noise equivalent of α_{in} . It should be noted that this has to be larger than the interface noise parameter, this is because if only a small amount of a phase is present then the double well potentials within the system will reduce this amount eventually to zero. There is a critical amount of phase i (depending upon the undercooling of the system) which must be nucleated in order to prevent this. As with the interface noise, when a small amount is added to phase i in the form of nucleation noise the same amount has to be subtracted from one of the other phases present in order to maintain Equation (6).

4 Results

There are two sets of results presented here, firstly results using only one solid phase. While these results can be produced more efficiently using a single phase solver these tests were carried out to show that the model works adequately in a system containing only one solid phase, they also show the anisotropy and interface noise working as desired. The second set of results show a series of simulations involving eutectics.

4.1 Two Phase Results

The first of these results shows a dendrite with four fold symmetry, the plot is the solute field. One of the assumptions used implicitly during the derivation of the anisotropic solver was that the anisotropic function η has to have 180° rotational symmetry, Figure 1 clearly shows this. Initially the side arms grow perpendicular to the local solute contours, this is almost perpendicular to the primary arm, however because the anisotropy favors growth in directions separated by 90° these side arms will turn to be perpendicular to the primary arms. As these side arms approach the side arms of the neighboring primary arm they will be deflected towards the radial direction.

4.2 Multi Phase Results

The first of these results shows the nucleation noise term working to cause the eutectic spacing to become roughly one third of it's initial spacing. The results in Figure 2 are similar to those presents by Nestler and Wheeler [6] who carried out the nucleation manually rather than by a noise term. Figure 2 clearly shows eutectics splitting to form narrower eutectics. If nucleation noise is left turned off then the results shown by Nestler and Wheeler [6] are recovered. As stated, the nucleation in this simulation can be carried out manually without the addition of a nucleation noise term. It is possible to envisage a system in which nucleation would occur but is sufficiently more complicated as to make manual nucleation too difficult. Figure 3 shows the results of a simulation without nucleation noise, the initial conditions were a circular region of solid, split into alternating bands of the two solid phases. Figure 4 is the same as the previous example however in this simulation the nucleation noise has been turned on, if this were to be done using a manual nucleation the there would have to be nucleation events corresponding to each of the transverse α - β pairs.

Figures 5 and 6 show a variation on a standard eutectic. To create these results a standard eutectic simulation was carried out however anisotropy was included and the liquid concentration was moved 0.5% off eutectic composition, this resulted in one of the eutectic phases being 'cut off' by the other and this remaining phase continueing to form a dendritic cell microstructure. Without anisotropy this would produce a significantly different result, rather than a cell type microstructure after the second phase is cut off the first phase would grow with a roughly flat interface. It should also be noted that in both of these cases nucleation noise is turned off. The differences between Figures 5 and 6 can be accounted for by anisotropy strength and initial eutectic spacing. Figure 6 also shows direct competition between adjoining dendritic arms and elimination of some of these arms.

References

- [1] J. Warren and W. Boettinger. *Acta. Metall. Mater.*, 1995, 43, pp. 689-703.
- [2] A. Wheeler, B. Murray and R. Schaefer. *Physica D*, 1993, 66, pp. 243-262.
- [3] R. Braun and B. Murray. *J. Crystal Growth*, 1997, 174, pp. 41-53.
- [4] Y. Kim, N. Provatas, N. Goldenfeld and J. Dantzig. *Physical Review E*, 1999, 59, pp. 2546-2549.
- [5] B. Nestler and A. Wheeler. *Comp. Phys. Comm.*, 2002, 147, pp. 230-233.
- [6] B. Nestler and A. Wheeler. *Physica D*, 2000, 138, pp. 114-133.
- [7] B. Nestler, A. Wheeler, L. Ratke and C. Stöcker. *Physica D*, 2000, 141, pp. 133-154.
- [8] I. Steinbach, F. Pezzola, B. Nestler, M. Seeßelberg, R. Prieler, G. Schmitz and J. Rezende. *Physica D*, 1996, 94, pp. 135-147.
- [9] J. Tiaden, B. Nestler, H. Diepers and I. Steinbach. *Physica D*, 1998, 115, pp. 73-86.
- [10] K. Jackson and J. Hunt. *Trans. Metall. AIME*, 1966, 236, pp. 1129-1142.
- [11] B. Wei, D. Herlach, F. Sommer and W. Kurz. *Mat. Sci. & Eng. A*, 1993, 173, pp. 357-361.
- [12] R. Goetzinger, M. Barth and D. Herlach. *J. Appl. Phys.*, 1998, 84, pp. 1643-1649.

5 Figure captions

Figure 1: A simulation demonstrating the use of four fold anisotropy and interface noise

Figure 2: Initially unstable, wide eutectics dividing to form narrower eutectics by way of nucleation noise

Figure 3: A simulation starting from a complex geometry with nucleation noise turned off

Figure 4: A demonstration of a complex geometry in which manual nucleation would be prohibitively complicated

Figure 5: An off eutectic composition showing the formation of dendritic cells and the use of anisotropy in a three phase system

Figure 6: An off eutectic composition showing the formation of dendritic cells and competition between these cells











