Thermoelectric Magnetohydrodynamics in Dendritic Solidification

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Declaration

I certify that this work has not been accepted in substance for any degree, and is not concurrently submitted for any degree other than that of Doctor of Philosophy (PhD) of the University of Greenwich. I also declare that this work is the result of my own investigations except where otherwise stated.

Andrew Kao (Author) Prof. Koulis Pericleous (Supervisor)

Dr. Valdis Bojarevics (Supervisor) Prof. Peter Lee (Supervisor) I would like to dedicate this thesis to my family and friends.

Abstract

The focus of this work is to investigate the effects of applying an external magnetic field to a solidifying liquid metal melt. The principle is that thermoelectric currents that are naturally inherent to solidification processes will interact with this magnetic field, resulting in a Lorentz force. This force will exist in a microscopic region in the vicinity of the solidification front, generating microscopic fluid flow in the liquid region which can significantly effect the mechanism of dendritic growth. The work contained in this thesis provides an initial insight into the complex behaviour of this process, through the use of numerical models.

To model the soldification dynamics, an enthalpy based model for dendritic growth in a supercooled melt is used in 2-dimensions and extended into 3-dimensions. The dendrite is defined as being equiaxed in nature and, for purely diffusion driven growth, numerical calculations show a good agreement with other methods under similar growth parameters. To investigate the effects of fluid dynamics, dendritic growth is tested under forced convection conditions and significant morphological changes occur. The incident tip velocity is increased and the downstream tip velocity is decreased; in agreement with many other authors investigating similar situations. In the presence of a magnetic field the Lorentz force will form in planes perpendicular to the direction of the magnetic field. Due to the morphology and anisotropy of the surface temperature, the nature of the flow is dependent on the relative orientation of the magnetic field and the crystallographic orientation of the lattice. Using a low magnetic field strength approximation, thus removing the non-linear and resistive terms in Navier-Stokes equation, the resulting fluid velocity is arbitrarily small so that convective transport is negated. At some time, when the morphological features of a dendrite are apparent, steady state simulations show the flow fields that exist with different orientations of the magnetic field. The results are compared to an analytic solution for the Lorentz force, which is described by reducing the morphology of a dendrite to a sphere and assuming that the surface temperature is equivalent to the anisotropy in the surface energy.

When the thermoelectric currents are large and the magnetic field strength is substantial the convective transport, non-linear and resistive terms become significant. The problem is purely 3-dimensional and it is shown that classical 2-dimensional boundary conditions lead to stagnant conditions. A 2-dimensional quasi 3-dimensional approximation is proposed and, with the magnetic field orientated in the (001) direction, the effect of heat and solute redistribution through convection on the crystal morphology is modelled. Two significant morphological changes occur; the first is a deflection of the dendrite tip and the second is the initiation of secondary branching into the incident flow. The deflection is caused by circulations at the tips of the dendrite; the circulations continuously provide a region of higher free energy on the incident side while lowering it on the other. The net effect is a bias of growth in the direction of incident flow. The increase in secondary branching, in a similar fashion to the deflection, is caused by both a circulation at the tip and also a global circulation around the entire dendrite, destabilising the incident interface and initiating secondary growth. To qualify the quasi 3-dimensional approximation, a moving mesh technique is developed that tracks a single tip of 3-dimensional growth and the similar morphological features are observed in comparison to the quasi 3-dimensional case.

Finally a discussion into possible extensions of this work is proposed and preliminary results for grain growth in the presence of a magnetic field are given.

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Nomenclature

Symbol	Description	Unit
A	area	m^2
A_0	current scaling factor	А
A_l^m, B_l^m, C_m, S_m	coefficients of solution to Laplace equation	-
$\alpha_0, \alpha_1, \alpha_2, \alpha_3$	anisotropic coefficients	Nm^{-1}
$lpha_k$	thermal diffusivity	$\mathrm{m}^2\mathrm{s}^{-1}$
В	magnetic field	Т
C	concentration	-
C_0	initial concentration	-
c_p	volumetric specific heat	$\mathrm{Jm^{-1}K^{-1}}$
\dot{D}	mass diffusivity	$\mathrm{m}^{2}\mathrm{s}^{-1}$
ϵ_4	anisotropic strength	-
\mathbf{E}	electric field	Vm^{-1} -
\mathbf{F}	force	Ν
f	liquid fraction	-
$\gamma\left(heta,\phi ight)$	surface energy	Nm^{-1}
γ_0	average surface energy	Nm^{-1}
$\Gamma\left(heta,\phi ight)$	surface stiffness	Nm^{-1}
H	volumetric enthalpy	Jm^{-3}
i, j, k	Cartesian array indicies	-
J	current density	Am^{-2}
k	partition coefficient	-
K	thermal conductivity	$\mathrm{Wm^{-1}K^{-1}}$
K(f)	step resistance term	Nm^3
κ	mean curvature	m^{-1}
L	volumetric latent heat	Jm^{-3}
L_0	characteristic length	m
Λ	interface	-
m_0	mass scaling factor	kg
m_L	liquidus slope	K

μ	dynamic viscosity	Pas
$\mu\left(heta,\phi ight)$	kinetic mobility	$\rm mKs^{-1}$
\hat{n}	normal unit vector	-
Ω	sub domain	-
ω	vorticity	s^{-1}
p	pressure	Pa
$P_m^l \cos\left(\theta\right)$	associated Legendre polynomials	-
Pe	Péclet number	-
Pr	Prandtl number	-
φ	conserved variable	-
Ψ	electric potential	V
q	charge	\mathbf{C}
\hat{Q}	surface heat flux	$\mathrm{Jm}^{-2}\mathrm{s}^{-1}$
Ŵ	volumetric heat production	$\mathrm{Jm^{-3}s^{-1}}$
$r, heta, \phi$	spherical polar coordinates	m
R_{tip}	tip radius	m
r_c	contact resistance	$\Omega \mathrm{m}^2$
Re	Reynolds number	-
ρ	density	$\rm kgm^{-3}$
S	Seebeck coefficient	VK^{-1}
ΔS	Seebeck power	VK^{-1}
σ	electrical conductivity	Sm^{-1}
t	time	S
Δt	time step	S
t_0	time scaling factor	S
$ au_0$	characteristic time	S
T	temperature	Κ
T_0	temperature scaling factor	Κ
T_{f}	temperature of fusion	К
T_m	equilibrium temperature	Κ
$\hat{ au}$	tangential unit vector	-
u	fluid velocity	ms^{-1}
u, v, w	x, y, z components of fluid velocity	ms^{-1}
$\mathbf{u_e}$	electron drift velocity	ms^{-1}
$\mathbf{u_{mesh}}$	moving mesh velocity	ms^{-1}
$\mathbf{u}_{\mathbf{ss}}$	steady state flow velocity	ms^{-1}
$\mathbf{u_{tip}}$	tip velocity	ms^{-1}
ν	interface velocity	ms^{-1}
V_C	concentration potential	-
V	volume	m^3
x,y,z	Cartesian coordinates	m

x_0	length scaling factor	m
Δx	cell size	m

Chapter 1 Introduction

Throughout history the study of metallurgy has been a prerequisite for the development of industry and it has been well understood that the combination of several elements provides a large variety of materials with varying physical properties. These mixtures are known as alloys and are widely used in the modern age; from mass produced steel to high performance materials such as Nickel based super alloys. Advances in optical and electron microscopes have shown that both pure metals and alloys solidify as crystalline structures known as dendrites. Moreover the formation and interaction of many dendrites known as the microstructure plays a significant role in the material properties. This has motivated the development of techniques to alter and control the microstructure formation. This can be achieved in the latter stages of solidification through the process of forging, heat treatment and even the use of ultrasonics. Another approach is to directly effect the dendritic growth during the solidification process, where it has been shown that the introduction of fluid motion (convection) can influence the formation of dendrites. Convection can be driven in the melt through natural buoyancy, pouring and through the use of electromagnetic fields. The mechanism for electromagnetic processing of materials is based on the interaction of the magnetic field with current giving rise to Lorentz forces which is the driver of convection. In some processes a rotating or oscillating field is responsible for generating current through electromagnetic induction. In other processes, such as the Vacuum Arc Remelting process, the magnetic field is a consequence of the large current present. The use of an external static magnetic field has received increased attention as advances in superconductivity have led to the viability of using high magnetic fields in industrial scale manufacturing processes.

The thermoelectric effect is the conversion of thermal energy to electrical energy and vice-versa; its effects have been demonstrated on the macroscopic scale in the form of refrigeration and power generation. However this theory is also applicable to the microscopic scale and is a naturally occurring phenomenon in dendritic growth. Therefore solidification in the presence of an external magnetic field may generate magnetohydrodynamic convection through the interaction of thermoelectricity and the magnetic field. This could lead to an additional control system to tailor the microstructure in such a way that it favours specific material properties. However, currently, the consequences of applying a magnetic field to dendritic solidification are not well understood.

1.1 Thesis Overview

There is a clear need to further the understanding of the fundamental effects of applying a magnetic field to dendritic solidification. The work contained in this thesis focuses on the change to the microstructure caused by thermoelectric magnetohydrodynamic fluid flow.

Experimentation can be a costly process and without a clear understanding of the underlying mechanics, experimental parameters can be difficult to predict. The use of numerical techniques is now common practice to provide initial estimates. In this work a numerical model is described that couples all of the physical mechanisms present together and predicts the resulting changes to the microstructure. The results provide an initial insight into the complex nature of the problem and highlight possibilities for controlling the development of the microstructure through the use of external magnetic fields.

1.2 Thesis Contributions

Although the concept of combining the fields of thermoelectricity and magnetohydrodynamics has existed for many years, applying this to dendritic growth specifically has thus far been relatively unexplored. This has provided the motivation for this work and the subsequent chapters attempt to answer the following questions:

What are the effects on microstructural evolution due to an externally applied magnetic field during dendritic solidification?

What is the mechanism that causes change to dendritic morphology?

To answer these questions time-dependent numerical models in both 2-dimensions and 3-dimensions are described for each of the three fundamental aspects: solidification, thermoelectricity and magnetohydrodynamics. The models are then coupled in different configurations providing an insight into how each individual aspect contributes to the overall process. The fully coupled transient model incorporates all of these aspects including solidification: thermal and solute transport, solutions for the electric potential, calculations for the thermoelectricity and Lorentz forces and the solution to the magnetohydrodynamic flow. In all cases the evolution of dendritic growth for both a single crystal and a grain is tracked.

The results provide an initial understanding of the complex nature of the system and provide a qualitative explanation for some of the experimentally observed changes. This thesis explores how varying key parameters affects the system and relates these to real materials such as the commercially used alloy Aluminium Silicon, which will be consistently used as an example throughout this work. A theoretical validation of the numerical results is provided through an analytic solution on a sphere.

1.3 Thesis Outline

Chapter 1 provides a brief introduction into fields of thermoelectricity, magnetohydrodynamics and metallurgy and describes the motivation for investigating such phenomena in the drive to control microstructural formation. This is followed by an overview of the work contained in this thesis and the contributions made in addressing the main research question. This chapter also contains a chapter by chapter outline of this thesis.

Chapter 2 incorporates background theory for the three fundamental fields that are being coupled in this work. This is intended as a summary, to highlight the necessary physics from each of the fields that is required when investigating this problem.

Chapter 3 reviews the current literature available and, since there is relatively little literature that directly investigates the effect being studied in this thesis, the review also branches into fields that are somewhat similar albeit not directly related. The review is split into four topics, dendritic growth, dendritic growth and convection, thermoelectric magnetohydrodynamics and dendritic growth in the presence of a magnetic field.

Chapter 4 describes the development of the numerical model used in simulating the different aspects of this research. The dimensionless model and the attainment of base SI scaling units are described. The numerical techniques for dendritic growth, thermoelectricity and magnetohydrodynamics are described in their discretised form along with their implementation. A quasi 3D approximation is developed to allow for some 3D features to be simulated in 2D and a moving mesh algorithm, which allows for full 3-dimensional simulations to be carried out, is proposed. The complete algorithm and the coupling between the various dynamics is described. Finally an appreciation is given to the numerical error by investigating approximations taken physically and numerically.

Chapter 5 presents the 2-dimensional and 3-dimensional results obtained through implementation of the numerical model. The first results investigate diffusion driven growth and the effect of anisotropic strength. Forced convection is then introduced to compare against other work that has been carried out in this field. Low magnetic field flow results are then presented to provide an insight into the fluid dynamics that will form through thermoelectric magnetohydrodynamics; these results are effectively diffusion driven as the convection does not alter the crystal morphology. Finally, fully coupled results for moderate to high magnetic field cases are presented.

Chapter 6 solves an analytic solution for the Lorentz forces on a sphere, where the surface energy of a dendrite is mapped onto the sphere. The numerical results are compared to the analytic solution to provide some form of theoretical validation of the numerical model.

Chapter 7 contains the conclusions of the predictions made in this research and discusses possible ideas for future work.

Appendix A details some of the less obvious analytic derivations that are somewhat tedious to incorporate in the relevant chapter, but are nevertheless important and are therefore included for completeness.

Appendix B lists publications from the author of this thesis that have been based on this research.

Chapter 2

Theory

2.1 Overview

In this chapter the fundamentals of thermoelectricity, magnetohydrodynamics and solidification are discussed. Coupling these three fundamentally different fields together is currently somewhat of a rarity and so to engage a wider audience a brief overview of the underlying principles and how they are related is presented.

2.2 The Thermoelectric Effect

The concept of the thermoelectric effect is attributed to Jean Charles Athanse Peltier and Thomas Johann Seebeck, who discovered this effect independently in 1821. Thermoelectricity is essentially the conversion of thermal energy into electrical energy, the fundamentals of which are detailed in semi-conductor physics. There are two necessary conditions. The first is a temperature difference along the liquid-solid interface and the second is a difference in absolute thermoelectric power (commonly known as the Seebeck coefficient) across the liquid-solid interface. The Seebeck coefficient is related to electron affinity, which is a measure of a materials ability to hold onto electrons. For a uniform composition under a thermal gradient electrons will migrate to the cooler region leaving behind a hole in the hot region; this occurs until an electrostatic equilibrium is formed. The



Figure 2.1: Electrons are promoted in the hot region and migrate to the cooler region until an electrostatic equilibrium is formed.

Seebeck coefficient is a relative quantity and in practice it is difficult to measure the absolute value. It is common to relate the Seebeck effect to the Seebeck Power, which is derived from the difference between Seebeck coefficients of the liquid and solid phases. When two materials are placed in thermal contact the material with a relative positive Seebeck coefficient acts as a p-type semiconductor and electrons pass to the material with a relative negative Seebeck coefficient, which acts as an n-type semiconductor. Electron affinity is both material and temperature dependent; for the latter it can be assumed that the solid and liquid phases will generally have different temperatures, thus a non-zero Seebeck Power will exist. When looking at solidification of supercooled pure materials, the temperature difference across the interface creates a significant Seebeck Power. For alloy solidification the material can be chosen such that a Seebeck Power exists irrespective of the temperature difference across the interface. However for an isothermal interface this exchange will instantaneously occur and reach a static equilibrium. It is also important to note that in most cases lower temperature materials generally have a positive Seebeck number.



Figure 2.2: Electrons migrate across the interface when two materials with varying Seebeck coefficient are placed in thermal contact. For an isothermal temperature T_I this exchange will be constant across the interface.

For non-isothermal interfaces the n-type material electrons will move from regions of higher temperature and follow the negative thermal gradient to cooler regions. This creates an imbalance in the distribution of charge and therefore electrons in the p-type material will travel in the opposite direction. This has the effect of electron-hole pairs forming in the hot region and recombining in the cooler region. Given that the current density (\mathbf{J}) is defined as the movement of positive charges, then a circulation of current forms; this is known as the Seebeck effect.

As more energetic electrons are moving from hot to cold and less energetic electrons are moving from cold to hot, there is a net movement of energy from the hot to cold regions and in the absence of any thermal sources both regions will eventually reach a thermal equilibrium. This converse effect is described by the Peltier and Thompson effects and is essentially the conversion of electrical energy into thermal energy and will be discussed later. To quantify the Seebeck effect Ohm's law for moving conductors is generalised to include a thermoelectric



Figure 2.3: Electrons and holes split at the hot region then diffuse along the negative thermal gradient recombining at the cooler region, the net effect is a circulation of current

term [1]:

$$\frac{\mathbf{J}}{\sigma} = \mathbf{E} + \mathbf{u} \times \mathbf{B} - S\nabla T \tag{2.1}$$

setting:

$$\Psi_{sb}\left(T\right) = \int SdT$$

integrated from some datum temperature. If S is constant away from the interface then:

$$\Psi_{sb} = ST$$

Introducing the electric potential 2.1 becomes:

$$\frac{\mathbf{J}}{\sigma} = -\nabla \left(\Psi_E + \Psi_{sb} \right) + \mathbf{u} \times \mathbf{B} = -\nabla \Psi + \mathbf{u} \times \mathbf{B}$$
(2.2)

For the situation given in figure 2.3, current flow can be described by the influence of a Seebeck potential difference:

$$\Delta \Psi = \oint \frac{\mathbf{J}}{\sigma} \cdot dl \tag{2.3}$$

Assuming at the interface there is a no-slip condition $(\mathbf{u} = 0)$ and preforming the integration around an infinitesimally thin loop:

$$\Delta \Psi = \Psi_E^i - \int_{T_H}^{T_C} S_A \nabla T \cdot dl - \Psi_E^i + \int_{T_H}^{T_C} S_B \nabla T \cdot dl = \int_{T_H}^{T_C} (S_B - S_A) \, dT = \int_{T_H}^{T_C} \Delta S \, dT \tag{2.4}$$

When the Seebeck coefficients are considered to be constant per material then the potential difference becomes:

$$\Delta \Psi = \Delta ST \tag{2.5}$$

Conservation of current gives:

$$\nabla \cdot \mathbf{J} = 0$$

$$\nabla^2 \Psi = \nabla \cdot \mathbf{u} \times \mathbf{B}$$
(2.6)

To solve for Ψ the problem can be split along the interface into two independent problems where 2.6 applies away from the interface and 2.5 applies to the interface. At the interface 2.5 can be split:

$$\begin{split} (\Psi_A-\Psi_B) &= \Delta ST \\ \Psi^i_A &= +\frac{1}{2}\Delta ST + \Psi^i_E(T) \\ \Psi^i_B &= -\frac{1}{2}\Delta ST + \Psi^i_E(T) \end{split}$$

The factor of $\frac{1}{2}$ represents the relative splitting of electron - hole pairs. From 2.4 $\Delta \Psi_E^i = 0$, however when separating the boundary at the interface to represent the

potential instead of the potential difference it is important to reintroduce $\Psi_E^i(T)$ as it accounts for time independent bound charges and is not simply a potential gauge as it is necessary to satisfy continuity of $\mathbf{J}\hat{n}$ at the interface. For a closed system any static charges will form in the corresponding hot and cold regions where the current circulates. Taking material B to be the solid and material A to be the liquid in a solidification problem, the domain can be written as two non-overlapping sub domains Ω_i :

$$\Omega_A \cup \Omega_B = \Omega$$
$$\Omega_A \cap \Omega_B = \emptyset$$
$$\Lambda = \partial \Omega_{AB}$$

Where the position of $\partial \Omega_A$ for an idealised system is at ∞ . The solution for Ψ



Figure 2.4: Domain decomposition splitting the problem into two independent problems with separate boundary conditions

is given by:

$$\frac{\partial \Psi}{\partial n} = 0 \quad \text{on} \quad \partial \Omega_A
\nabla^2 \Psi = \mathbf{u} \times \mathbf{B} \quad \text{in} \quad \Omega_A
\frac{\partial \Psi}{\partial n_A} = \frac{\partial \Psi}{\partial n_B} \quad \text{on} \quad \Lambda
\Psi = +\frac{1}{2}\Delta ST + \Psi^i_E(T) \quad \text{on} \quad \Lambda_A
\Psi = -\frac{1}{2}\Delta ST + \Psi^i_E(T) \quad \text{on} \quad \Lambda_B
\nabla^2 \Psi = 0 \quad \text{in} \quad \Omega_B$$
(2.7)

A good example that will be used consistently throughout this thesis is the binary alloy Aluminium Silicon (AlSi), for the temperature regime of interest Aluminium and Silicon have a Seebeck Power of $S \sim 10^{-3} \frac{V}{K}$.

As energetic electrons passes through the conductor energy is transported and dissipated locally through joule heating. However for a conducting material that has a Seebeck coefficient that varies with temperature an extra term is added to the joule heating equation. This effect is known as the Thompson effect and was discovered in 1841 by Lord Kelvin. If a current is passed through this material the heat production per unit volume (W) is given by:

$$W = \frac{\mathbf{J}^2}{\sigma} - \mathbf{J}T \cdot \nabla S \tag{2.8}$$

where the first term on the right is the effect of joule heating and the second term is the change due to the Thompson effect. For a pure material, where the medium is of uniform composition and the Seebeck Coefficient is only a function of the temperature the Thompson term may be re-written giving:

$$W = \frac{\mathbf{J}^2}{\sigma} - T \frac{dS}{dT} \mathbf{J} \cdot \nabla T \tag{2.9}$$

A similar effect also occurs at an interface where the Seebeck coefficient varies discontinuously. At this location the Peltier effect accounts for transport of heat through the splitting or recombining of electron-hole pairs and can be described as a local surface heat flux (Q) in terms of the normal component of current density $(\mathbf{J}\hat{n})$:

$$Q = r_c \mathbf{J}^2 \hat{n} + \Delta ST \mathbf{J} \hat{n} \tag{2.10}$$

where the first term is the joule heating at the interface, r_c is the contact resistance and the second term is the Peltier effect. The Seebeck power appears in the Peltier term and the sign provides the net movement of energy. For closed systems in the absence of any thermal sources this effect will drive the system to become isothermal. However for materials that exhibit thermoelectric properties external currents can be used to re-distribute thermal energy. This is the concept behind Peltier coolers such as electric fridges which do not require the expansion of Freon. In both cases the joule heating term is thermodynamically irreversible, while the Peltier and Thompson effects are theoretically reversible. The purpose of this work is to investigate the magnetohydrodynamics that interact with the current producing Seebeck effect and so joule heating and the Peltier and Thompson effects have been neglected. However a quantification of their relative magnitudes using typical values for supercooled dendritic solidification and hypothetical predictions of the morphological changes caused by them are discussed in the future work section.

2.3 Magnetohydrodynamics

Magnetohydrodynamics (MHD) is a relatively modern subset of classical fluid dynamics; initiated by Hannes Alfvén in 1942 [2]. The basic principle is that magnetic fields can induce or interact with currents in a conducting fluid; generating forces accelerating the fluid. For induced currents this can lead to a change
in the magnetic field itself. Electromagnetic fields in material processing have now become widely used across a broad spectrum of processes. The most common usage is magnetic stirring, where an imposed oscillating or rotating magnetic field induces an electric current inside the liquid metal bringing about convection. However in some cases the generation of magnetohydrodynmic flow is a by product of the actual process. In some processes a large voltage is imposed between an electrode and the liquid melt, this introduces a large current, which induces a magnetic field which interacts creating Lorentz forces and fluid motion. For the purpose of this work the external magnetic field is considered to be sufficiently large that the changes from any induced currents will be insignificant. The current will be formed through the thermoelectric effect described in the previous section and in the presence of a magnetic field this will generate a Lorentz force in terms of charge q:

$$\mathbf{F} = q \left[\mathbf{E} + (\mathbf{u}_{\mathbf{e}} \times \mathbf{B}) \right] \tag{2.11}$$

Combining this with the definition for electrical current and integrating over an infinitesimally small volume gives the volumetric electrical current or current density. For this work \mathbf{J} is the thermoelectric current describer in the previous section:

$$\frac{\mathbf{F}}{V} = \mathbf{J} \times \mathbf{B} \tag{2.12}$$

Thus the equations that describe MHD flow of this kind are the classical Navier-Stokes equations with the Lorentz force appearing on the right hand side:

$$\rho \frac{\partial \mathbf{u}}{\partial t} + \rho \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{J} \times \mathbf{B} - K(f)$$
(2.13)

The final term on the right represents a step resistance term, that effectively prevents flow from entering a solid region by introducing a resistive force that exactly balances the driving forces at the interface. The final condition on the solution for 2.13 is continuity:

$$\rho \nabla \cdot \mathbf{u} = 0 \tag{2.14}$$

2.4 Hypo-Eutectic Solidification

The simplest alloy consists of two elements, which when mixed form a binary alloy. How the alloy solidifies depends on the initial temperature and concentrations of each component of the alloy. For eutectic solidification the concentrations are selected such that both the alpha and beta phases solidify at the same temperature, this is known as the eutectic point. The phase diagram for a standard binary alloy is given in figure 2.5. If the initial position is selected such that alpha is the primary phase, i.e. left of the eutectic point then hypo-eutectic solidification will occur, conversely if beta is the primary phase then hyper-eutectic solidification occurs. Under eutectic conditions bands of alpha and beta form alternatively. In this work beta phase solidification is not considered and thus microstructural evolutions of hyper-eutectic and eutectic solidification are neglected. Aluminium Silicon alloys are widely used in industrial applications due to the ease of casting, tensile strength and light weight. The most common application is its use in aeronautics. Combined with excellent thermoelectric properties this makes AlSi alloys an excellent candidate for this work and thus continuing from the example in the thermoelectric section, the phase diagram of AlSi in figure 2.6 is similar to that of a classical binary alloy, with the exception that Aluminium has zero



Figure 2.5: Phase diagram of a binary system

solid solubility in silicon at any temperature and so the beta phase is pure Silicon. During hypo-eutectic solidification of an AlSi alloy the alpha phase will solidify first as almost pure aluminium dendrites. Aluminium has a face centred cubic lattice structure and with a small amount of Silicon, the Aluminium lattice structure will be dominant exhibiting four-fold symmetry or equiaxed dendrites, with a boundary layer of pure Silicon forming close to the interface.

In this work an enthalpy based method is used to calculate the evolution of the crystal morphology, an explanation for this choice is given later and for the purpose of this review the equations are written in terms of enthalpy. The liquid fraction (f) describes the volumetric proportion of solid to liquid over a given volume. f is defined as f = 0 is solid and f = 1 is liquid, intermediate values are partially solid and represent the mushy zone, f can be considered as a measure of



Figure 2.6: Phase diagram of part of Aluminium Silicon system

how ordered the atoms are to the lattice or as a non-dimensional entropy figure 2.7 provides a qualitative representation of this. The interface is defined as f = 0.5, for material properties that are continuous at the interface it is assumed f varies linearly, while for material properties that are discontinuous at the interface f represents a step function. To relate f to enthalpy, the volumetric enthalpy (H) is defined as:

$$H = c_p T + fL \tag{2.15}$$

For binary alloys the idea developed by Crowley *et al.* [3] is used, where the concentration potential (V_C) is defined as:

$$V_C = \frac{C}{f(1-k) + k}$$
(2.16)



Figure 2.7: The liquid fraction can be thought of as a function of entropy

The partitioning coefficient defines the relative contributions to the phases of materials A and B during solidification and is defined by:

$$k = \frac{C_s}{C_l} \tag{2.17}$$

In the case of pure materials k = 1. The conservation of enthalpy is given by:

$$\frac{\partial H}{\partial t} = \nabla \cdot (K\nabla T) - \nabla \cdot (\mathbf{u}H) \tag{2.18}$$

The thermal conductivity (K) is assumed to vary linearly between K_s in the solid to K_l in the liquid. The conservation of solute is given by:

$$\frac{\partial C}{\partial t} = \nabla \cdot (D\nabla V) - \nabla \cdot (\mathbf{u}C) \tag{2.19}$$

This equation is similar to that of conservation of enthalpy except the mass diffusivity between the solid and liquid is significantly different. A good approximation is to use the Scheil assumption $D_s = 0$. The equilibrium solidification temperature (T_m) is given by:

$$T_m = T_f + m_L C_0 \tag{2.20}$$

The first term is the temperature of fusion, the second term is the change in solidification temperature due to the initial solute concentration as seem in the phase diagram from figure 2.5, the liquidus slope m_L is assumed to be constant and for pure materials $m_L = 0$. However due to the curvature, crystal anisotropy, tip velocity, solute partitioning and redistribution the interface is undercooled to the temperature T^i :

$$T^{i} = T_{m} - \frac{\Gamma\left(\theta,\phi\right)}{L} T_{m}\kappa - m_{L}\left(C_{0} - C_{l}^{i}\right) - \frac{\nu}{\mu\left(\theta,\phi\right)}$$
(2.21)

The second term accounts for the crystal anisotropy and local interface curvature and the third term accounts for the variance in solute concentration and the final term accounts for kinetic effects. The mean curvature can be defined by:

$$\kappa = \frac{\kappa_1 + \kappa_2}{2} = \frac{1}{2} \nabla \cdot \frac{\nabla f}{|\nabla f|} \tag{2.22}$$

Neglecting kinetic effects consider a pure solidifying sphere, where there is no preferential direction of growth ($\gamma = const$), then in the absence of any random fluctuations the free energy will be a linear function of the local curvature, which is constant at any point on the interface of a sphere. However if we now include the lattice structure, then there will be regions where the surface energy is lower due to the binding energy into the lattice. This gives a preferential direction of growth, which is known as the crystal anisotropy and is a 'macro' relationship to the binding energy required at different orientations to the crystal lattice. The anisotropy is therefore one of the key parameters which determines the overall crystal morphology by providing preferential direction of growth. The value for the anisotropy is material dependent and in practice difficult to measure.

2.5 Summary

In this chapter a basic overview of the three fundamental fields that are applied to this research are given. The origins and conditions for thermoelectric currents are discussed. Adaptations to Navier-Stokes equations to form the magnetohydrodynamics equations are given and the equations of solidification incorporating the anisotropy introduced through the crystal lattice are given.

Chapter 3

Literature Review

3.1 Overview

In this chapter a review of existing research is given. The review focuses on four main topics; the fundamentals of dendritic growth, current uses of magnetic fields in industrial applications, direct observation or modelling of thermoelectric magnetohydrodynamics and applying a magnetic field to a solidifying alloy.

3.2 Dendritic Growth

Solidifying metals form as crystalline structures known as dendrites, the shape of the dendrites can be related to the underlying crystallographic lattice and a large variety of shapes exists in nature [4]. The simplest crystallographic shape is when the metal solidifies with a cubic lattice and if the dendrite is unconstrained will preferentially grow in 6 orthogonal directions exhibiting 4-fold symmetry also known as equiaxed dendrites [5; 6].

Ivanstov [7] was the first to attempt to provide an analytic solution for supercooled dendritic growth, this work was later generalised by Horvay *et al.* [8]. A result from these studies was a solution relating the tip velocity and tip radius of the form:

$$u_{tip}R_{tip} = C \tag{3.1}$$

However to fully characterise the operating state of dendritic growth Oldfield [9] proposed through a stability argument the condition must also hold:

$$u_{tip}R_{tip}^2 = C \tag{3.2}$$

This work was later formalised and extended by Langer et al. [10]

In recent times sophisticated numerical models are used to predict the evolution of dendritic growth, the most common are cellular automata, front tracking, level set and phase-field.

Cellular automaton based methods [11–14] use a set of rules, where the rules are defined to be representative of the evolution. This can be based on the evolution of dendritic growth where, one approach is to use analytic solutions to dendritic growth to define the rules [11] [12], or can be based on the evolution of grains [13].

Front tracking methods, where the location of the interface is well known by either deforming the mesh such that the interface coincides with the mesh [15] or more commonly to use a Lagrangian approach overlaid onto some static background mesh. For the latter case the interface is displaced in time by satisfying heat balance for every interfacial node. To keep an accurate solution, points at the interface may need to be added and redistributed as the surface area increases. It is also necessary to relate the interfacial solution to the background mesh, this can be achieved by modification to the finite differencing scheme [16–18] or finite element scheme [19; 20] close to the interface to include the interfacial points, or by generating source terms on the background mesh through some form of interpolation from the solution of the interface [21].

In level set methods [22; 23] the level set function is usually defined as a distance function and is used to evolve the liquid solid interface though an advection equation. The interface velocity is defined such that it takes into account heat balance and interface undercooling. The most significant difference to front tracking methods is that as the interface is not exactly known and the interfacial conditions are not explicitly satisfied, but are assumed to be implicitly satisfied through the update of the level set function.

Phase-field methods [22; 24–26] use a diffuse interface approach which assumes the interface to have some finite width. The order parameter is prescribed to vary continuously across the interface and so are material properties that may vary between the liquid and solid. An energy function is defined across the diffuse interface which accounts for the interface effects and heat balance. By minimising the energy function in this region an evolution equation can be calculated, which is used to advance the interface. Phase-field methods if set up correctly can be very accurate, however it is necessary to have very small cell sizes in the vicinity of the interface, which then due to the large disparity in length scales necessitates the use of very sophisticated adaptive meshing techniques [27].

An alternative approach is to use the Enthalpy based method. First proposed by Tacke *et al.* [28; 29] and further developed by Voller *et al.* [30; 31]. The general principle is to take the order parameter described in a phase field method to be a level set function that can be used to directly calculate local interface curvature and velocity. This approach removes the necessity to minimise the energy equation as in phase field methods.

For this work the Enthalpy method has been chosen to model the phase change during solidification. This method was chosen for several reasons; the method directly accounts for local curvature and interface effects, the method can be more easily extended into 3-dimensions when compared to front tracking methods where it becomes quite complex and the method is fast compared to full phasefield methods.

Under stagnant conditions with no random fluctuations models of dendrites grow symmetrically, which is advantageous as it allows exploitation of these symmetry lines to increase the domain size. Karma *et. al* showed the effects of anisotropy on the crystal morphology, figure 3.1 shows isotropic dendrite growth. The solution should be spherical (if the seed is spherical), however a spherical solution is unstable and tiny perturbations, which in this case are introduced through the mesh deform the shape. Figure 3.2 shows growth with a high crystal anisotropy representative of face centred cubic materials.

Karma *et al.* [33] also investigated the effects of under cooling on dendritic growth. Figure 3.3 shows a result with moderately high anisotropy at low under coolings using a phase-field method.

Figure 3.4 shows a result from Tan *et al.* [34] who investigated the evolution of a Ni-Cu alloy under stagnant conditions using a finite element based level set approach. When using a 2-dimensional model to simulate similar situations the morphology will essentially represent a slice along one of the primary axes, such that four out of the six primary arms are captured.

Introducing convection into the melt has been shown to have a significant impact on the evolution of dendritic growth [35–54] and it has been suggested



Figure 3.1: Simulation of diffusion driven dendritic growth with isotropic surface energy from Karma et~al. [32]



Figure 3.2: Simulation of diffusion driven dendritic growth with isotropic surface energy from Karma et~al. [32]



Figure 3.3: Simulation of diffusion driven dendritic growth from Karma *et al.* [33]



Figure 3.4: Evolution of a Ni-Cu based alloy in stagnant conditions from Tan et al. [34]

that convection is the primary cause of differences between experimental results and prediction [55; 56]. Convection is almost always a naturally occurring phenomenon during solidification and is present through changes to density as the solid shrinks [57–59]. In processes where significant thermal gradients exist buoyancy driven flow can play a role in the final microstructure [35; 36; 52–54]. Melt convection can also be artificially introduced through gravitational forces and electromagnetic stirring, the latter of which will be discussed later. During earth bound electromagnetic levitation strong convective flows are a consequence of counter balancing the effects of gravity [60; 61]. Therefore there has been much interest in quantifying these effects experimentally and introducing melt convection into numerical models.

The first experimental study on the effects of convection on dendritic growth were preformed by Huang *et al.* [35; 36], where the study involved investigating the effect of buoyancy driven flow in succinonitrile dendritic growth. Gravitational forces introduced convection and it was shown that dendritic growth under these conditions had different tip velocities and morphological shape to diffusion drive growth.

Experiments have also been designed to investigate the effects of forced convection on Ammonium Chloride crystals by Ramani *et al.* [37] and Appolaire *et al.* [38]. By initiating growth in the upper regions of the solution the change in density causes free dendritic growth to take place as the dendrites settle under the influence of gravity. The heavier dendrites sink to the bottom and in the moving frame of the dendrite this provides a similarity to forced convection. The results show that the tip velocities were significantly altered when compared to diffusion driven growth. The crystals that maintained an alignment to the direc-

tion of flow experienced the largest morphological changes compared to rotating dendrites. The incident tips grew faster and become elongated, while the downstream tips experienced a reduction in velocity, an experimental result is given in figure 3.5.



Figure 3.5: Settling of Ammonium Chloride crystals under the influence of gravity [37]

Numerical models have since been designed to model the observed changes, this is achieved by extending the modelling techniques outlined previously and assuming that the solid phase is both rigid and stationary. This essentially causes only a single change to the solidification mechanics, which appears as the final term in the transport equations 2.18 and 2.19. In regions of high velocity this term becomes significant and the redistribution of both solute and temperature provides a change to the free energy at the interface influencing the growth. Incorporating a constant velocity boundary condition in 2-dimensions has been conducted with similar observable results. Figure 3.6 are results from Tong *et al.* [40], where the incident flow is directed on to the north tip and is forced around the solid dendrite region altering the dendrite morphology and the thermal boundary layer. Similar morphological changes have been modelled by Zhao *et al.* [42] and their results are shown in 3.7. In some cases depending on the incident velocity downstream vortices have been shown to form in 2-dimensions, an example of this from Al-Rawahi *et al.* [41] is given in figure 3.8.



Figure 3.6: Direction of velocity and thermal field in forced convection from Tong *et al.* [40]

Similar investigations have also been conducted in 3-dimensions [34, 43, 44, 45, 46], where the most significant difference is that the flow is able to pass over the dendrite and is not necessarily constricted to going around. The work carried out by Jeong *et al.* [43] is shown in figure 3.9 where similar morphological changes to the 2-dimensional models is observed and the flow can be seen to pass over and under the crystal arms. Tan *et al.* [34] investgated the influence of convection in



Figure 3.7: Solute concentration in forced convection from Zhao et al. [42]



Figure 3.8: Downstream circulations in forced convection from Al-Rawahi *et al.* [41]

Ni-Cu based alloys and the numerical results of this work are given in figure 3.10. Figures 3.11 and 3.12 show work carried out by Al-Rawahi *et al.* and Lu *et al.* respectively, where the morphological changes are similar. In all cases although the numerical parameters may vary, it is the same mechanism that is causing the changes to the crystal morphology.



Figure 3.9: 3D morphology and direction of velocity in forced convection from Jeong *et al.* [43]



Figure 3.10: 3D Ni-Cu alloy in a forced convection from Tan $et \ al. [34]$



Figure 3.11: Numerical simulation of dendritic growth in a forced convection at an early time and a well developed crystal from Al-Rawahi *et al.* [44]



Figure 3.12: 3D forced convection results from Lu et al. [46]

3.3 Solidification in the Presence of Magnetic Fields

Introducing convection into solidification process through the use of magnetic fields has been used extensively over many decades [62] and the governing equations were outlined in the previous section. Typically electromagnetic stirring is achieved through the use of AC or rotating magnetic fields, which induce currents in the solidifying melt. These currents in turn interact with any imposed and induced magnetic fields producing the Lorentz force in the melt accelerating the flow. The generation of strong macroscopic flows during solidification can have positive effects on the final material properties. The mixing caused by the fluid dynamics can provide homogenisation of the alloy elements which will reduce porosity and macro segregation, preventing cracks. The shear stress caused by the fluid dynamics could cause newly formed dendrites to break off resulting in a grain refinement [62–65].

In systems where large currents are present as part of the process, the self-

induced magnetic field can interact with the currents generating convection; the Vacuum Arc Remelting process (VAR) imposes a voltage between the electrode and the remelting ingot, this introduces a current which via joule heating causes the metal to remelt. This current in turn induces a magnetic field which interacts creating Lorentz forces and fluid motion and consequently the convective effects can alter the solidification process [66]. A similar situation also occurs in modelling the Electro-Slag Remelting process, to get an accurate result the induced magnetic field must also be considered [67; 68]. Another industrial process where induced MHD effects occur is inside the Aluminium cell, where again a large voltage exists between the anode and cathodes. The resulting induced magnetic field has been shown to bring about MHD flow causing rotating waves that could lead to an undesirable sloshing effect [69; 70].

When a DC field is placed across a solidifying melt where there are no electrical currents present then the only interaction is between any fluid velocity in the system and the magnetic field. This interaction will cause a dampening force on the flow and can be used to prevent or marginalise any unwanted effects from convection [71; 72]. However when currents are present in the system the static magnetic field will interact with these producing a driving Lorentz force, these currents can be artificially imposed or like thermoelectric currents may be an inherent part of the solidification process.

3.3.1 Thermoelectric Magnetohydrodynamics

In the previous section it was shown that convective effects can have a significant impact on dendritic growth. The purpose of this study is to investigate the effects of a special case of convection, generated through the interaction of thermoelectric currents and an external magnetic field. Although proposed by Shercliff [1] many years ago, the use of Thermoelectric Magnetohydrodynamics (TEMHD) as an industrially viable application has only recently started to receive serious consideration. So far the implementation of such techniques have been on the macroscopic scale and thus far it is been generally used for the purpose of a self stirring melt. In many industrial applications of liquid metals albeit casting, or plasma containment in nuclear fusion reactors a non uniform thermal field will form. Providing that the liquid metal is conducting this will inherently introduce thermoelectric currents and in the presence of a large enough magnetic field the Lorentz forces can become the predominant driving force of fluid motion. The fluid dynamics of natural convection were shown to be affected through this mechanism in a simplified model investigating the flow inside a cube of conducting liquid metal [73].

Work carried out by Zhang *et al.* [74] has shown that given a reasonable jump in Seebeck number of $20 \,\mu \text{KV}^{-1}$ with a moderate thermal gradient of 2.8K/cm that a permanent Neodymium rare earth magnet was sufficient to provide velocities in a conducting fluid of around 30mm/s. Another recent publication by Jaworski *et al.* [75] created a self stirring lithium melt and reported macroscopic velocities of the order of $30 \,\text{cms}^{-1}$, significantly high enough to produce intense stirring. Although the application of such techniques has not yet been implemented on an industrial scale, the increased scientific attention has broadened the spectrum of possible uses. For this work the same concept is being utilised, however the goal is to investigate the TEMHD in solidification on a microscopic scale.

This effect has also been modelled in 3-dimensions in the Zone melting process,

which is used in purifying crystals and thermoelectric currents naturally exist [76]. Investigations into using thermoelectric magnetohydrodynamics to control weld shapes has also been conducted and it was shown that the orientation of the magnetic field caused the weld shape to significantly change [77].

3.3.2 Dendritic Growth in the Presence of a Magnetic Field

The concept of using thermoelectric magnetohydrodynamics as a solidification tool was first proposed by Moreau *et al.* [78], who investigated the effects of a moderate magnetic field (up to 1.5T) on the microstructure of three different alloys: Bi-Sn (60% Bi), Cu-Ag (45% Cu) and Pb-Sn (not given) [79]. The Seebeck Power for each of these material pairings is $5 \times 10^{-5} \text{ VK}^{-1}$, 10^{-5} VK^{-1} and less than 10^{-6} VK^{-1} respectively. Each material is solidified with and without a magnetic field and for the materials with a higher Seebeck number it was found that the crystal morphology was significantly changed. With Pb-Sn which has electric and thermal conductivities similar to Bi-Sn, the magnetic field did not appear to have any noticeable effect on the crystal morphology. Evidence for the existence of microconvection is apparent by an increase in freckling of the fully solid alloy. The authors attribute this micro convection to the interaction of thermoelectric currents and the magnetic field. Simplified models have been devloped to investigate the fluid dynamics on dendrite like shapes [80] or using axi-symmetric approaches [81].

This specific field had not received much attention until recently, when several experimental papers were published [82–87], perhaps due to the improvement of superconducting magnets reducing the cost in designing experiments and allowing for higher magnetic fields to be used. Li *et al.* [83] report experimental results of Al-Cu alloy both 0.85% wt and 4.5% wt in the presence of a high magnetic field. The melt first undergoes directional solidification of rates between $1.5 \,\mu ms^{-1}$ and $100 \,\mu ms^{-1}$. To look at the evolution of the solid liquid interface the melt is withdrawn from the Bridgeman apparatus and immediatley quenched. The external magnetic field points in the direction of solidification and magnetic field strengths up to 10 T are applied. For the lower concentration alloy, with no magnetic field the liquid solid interface is planar. In the presence of a 10T magnetic field the interface destabilises to a cellular array. When the concentration of the alloy is increased to 4.5% weight a deflection parallel to the direction of the magnetic field is observed and secondary branching is increased. The authors attribute these changes to convection generated via the thermoelectric magnetohydrodynamic interactions.

Preliminary results in 2-dimensions [88–91] and in 3-dimensions [92], which will be presented later in this thesis provide a theoretical mechanism for some of these observed phenomena.

For very high magnetic fields it has been suggested that the resistive term $(\mathbf{u} \times \mathbf{B})$ becomes large enough to significantly damp out convection and that the Lorentz force acting inside the solid region becomes significant enough to shear the dendrites apart influencing the columnar to equiaxed transition [93].

3.4 Problem Description

To understand the underlying phenomena that take place during dendritic solidification in the presence of a magnetic field, an idealised situation is perceived. A small section of the liquid melt is considered in the bulk away from any wall. The liquid melt begins in a super-cooled meta-stable state, where the temperature is in the primary alpha regime just above the Eutectic line. A constant uniform magnetic field is applied across the whole domain. At some time nucleation of a small spherical isothermal seed occurs in the centre of the domain and hypo-eutectic solidification begins. The alpha phase is prescribed to be equiaxed dendritic growth and as the tips begin to form the crystal is no longer isothermal and thermoelectric currents circulate between the cold tips and the hot roots. In the presence of a magnetic field, these currents bring about Lorentz forces acting in the liquid phase driving MHD flow, this causes changes to the thermal and solutal transport mechanisms altering the available free energy and overall causing a change to the dendritic morphology.

3.5 Summary

This chapter provided a summary of the research conducted by other authors related to the focus of this thesis. A review of the attempts made both experimentally and numerically to understand the underlying fundamentals that occur during dendritic growth is presented, along with current uses and applications of magnetic fields in solidification processes. Examples where thermoelectric magnetohydrodynamic fluid flow has been physically observed or modelled and papers that have looked at applying a magnetic field to a solidifying alloy are also given.

Chapter 4

The Numerical Model

4.1 Overview

This chapter will discuss the numerical techniques used in modelling the phenomena described in the previous chapter. This involves recasting the equations in a dimensionless discretised form and to develop coupling between the dependent variables in each section of the model. The solution procedure for the solidification, electric potential and MHD flow are described separately for either a time independent situation or over a finite time step. The complete algorithm consisting of the solved order and the coupling mechanics is given at the end.

4.2 Dimensionless Numbers

To help with numerical stability a dimensionless form for all the equations is employed. A good example of why this is necessary is to consider the size of a single control volume; where $dx = 010^{-7}$ m leading to a volume of 010^{-21} m³, which for some parabolic numerical solvers may be considered to be zero. To over come this problem, scaling factors for all of the base SI units need to be found such that the values are neither too large or too small. Dimensionless terms are represented by the superscript *. A characteristic temperature scaling is obtained by:

$$T^* = \frac{T - T_f - m_L C_0}{\frac{L}{c}}$$

$$\frac{L}{c} = T_0$$
(4.1)

For the case of Aluminium $L \sim 4 \times 10^5 \,\mathrm{JK^{-1}}$. Close to the solidification temperature of Aluminium measurements by Buyco *et al.* [94] find $c \sim 1.2 \times 10^3 \,\mathrm{Jkg^{-1}K^{-1}}$. This gives a temperature scaling $T_0 \sim 3.3 \times 10^2 \,\mathrm{K}$. The thermal diffusivity α_k in general decreases with temperature and from the work by Kaschnitz *et al.* [95] a good value in this temperature range is $\alpha_k \sim 3 \times 10^5 \,\mathrm{m^2 s^{-1}}$. A characteristic time scale can be obtained by:

$$t^* = \frac{t}{t_0} = \frac{t}{\left(\frac{\alpha_k}{L}\right)^{\frac{1}{2}}}$$
(4.2)

The length scale is defined in terms of α_k :

$$x^* = \frac{x}{x_0} = \frac{x}{(\alpha_k t_0)^{\frac{1}{2}}} \tag{4.3}$$

The effect of scaling length, time and temperature in this way effectively set $\alpha_k^* = 1$. For aluminium the characteristic length scale is $x_0 \sim 5 \times 10^{-8}$ m and the characteristic time scale is $t_0 \sim 7.5 \times 10^{-11}$ s. Using a similar procedure the mass scale is derived by scaling the non-dimensional density $\rho^* = 1$:

$$\rho^* = \frac{\rho}{\rho_0} = \frac{\rho}{\frac{m_0}{x_0^3}}$$

$$m_0 = \rho x_0^3$$
(4.4)

This gives a mass scaling factor for Aluminium of $m_0 \sim 2.5 \times 10^{-19}$ kg. Finally by scaling the electrical conductivity to $\sigma^* = 1$ the scaling factor for the current becomes:

$$\sigma = \frac{t_0^3 A_0^2}{x_0^3 m_0} \tag{4.5}$$

$$A_0 = \frac{m_0 x_0^3 \sigma}{t_0^3} \tag{4.6}$$

In Aluminium $A_0 \sim 5 \times 10^{-2} \,\mathrm{A}$.

From work carried out by Galenko *et al.* [96] studying undercooled Ni-Zr alloys with similar undercoolings in this study the steady state tip velocities reported are in the region of $10 \text{ ms}^{-1} - 25 \text{ ms}^{-1}$. There is no detailed literature regarding Aluminium under these conditions, but assuming that this provides a reasonable estimate for Aluminium alloys this corresponds to a dimensionless growth velocity of $V^* = 0.016 - 0.04$.

4.3 The Enthalpy Method

Using the scaling factors described in section 4.2 the dimensionless form of the enthalpy method can be rewritten and equation 2.15 becomes:

$$H^* = T^* + f (4.7)$$

and the dimensionless interfacial temperature becomes:

$$T^{i*} = -\kappa \frac{\Gamma(\theta, \phi) T_m c_p}{L^2} - \frac{c_p m_L}{L} C_0 \left(1 - V_C\right) - \frac{c_p \nu}{L \mu\left(\theta, \phi\right)}$$
(4.8)

For clarity all variables from this point forward will be dimensionless unless otherwise stated and the superscript * will be removed. A uniform Cartesian finite difference mesh is used for solving the enthalpy method. For a given solid front with a known temperature and solute field, the evolution of the front is solved by discretising the solidification equations, the indices i, j, k represent the indices in the x, y, z directions respectively. Equation 2.15 becomes:

$$T_{i,j,k} = H_{i,j,k} - f_{i,j,k}$$
(4.9)

The concentration potential from equation 2.16 becomes:

$$V_{Ci,j,k} = \frac{C_{i,j,k}}{f_{i,j,k} + k(1 - f_{i,j,k})}$$
(4.10)

If a cell is 0 < f < 1 then it is considered to be part of the interface and by neglecting kinetic effects the interfacial temperature from equation 2.21 becomes:

$$T_{i,j,k} = -\kappa\gamma + m_L(1 - V_{Ci,j,k}) \tag{4.11}$$

where the mean curvature (κ) in 2.22 can be written in terms of the liquid fraction:

$$\kappa = \frac{(f_{yy} + f_{zz})f_x^2 + (f_{xx} + f_{zz})f_y^2 + (f_{xx} + f_{yy})f_z^2 - 2f_x f_y f_{xy} - 2f_x f_z f_{xz} - 2f_y f_z f_{yz}}{(f_x^2 + f_y^2 + f_z^2)^{\frac{3}{2}}}$$
(4.12)

the subscripts represent the derivatives in the corresponding directions:

$$f_x = \frac{\partial f}{\partial x} = \frac{f_{i+1,j,k} - f_{i-1,j,k}}{2dx}$$
$$f_{xx} = \frac{\partial^2 f}{\partial x^2} = \frac{f_{i+1,j,k} - 2f_{i,j,k} + f_{i-1,j,k}}{dx^2}$$

The angles θ and ϕ are the angles between the local normal and the x-axis, such that:

$$\phi = \tan^{-1} \frac{f_y}{f_x}$$
$$\theta = \tan^{-1} \frac{f_z}{(f_x^2 + f_y^2)^{\frac{1}{2}}}$$

For equiaxed crystals $\gamma(\theta, \phi)$ can be described by cubic harmonics. Originally proposed by Karma *et. al* [33] taking the harmonics to be orthogonal gives the form:

$$\frac{\gamma(\theta,\phi)}{\gamma_0} = (1-3\epsilon_4) \left[1 - \frac{4\epsilon_4}{(1-3\epsilon_4)} (N_x^4 + N_y^4 + N_z^4) \right]$$
(4.13)

where N_x , N_y and N_z represent unit vectors along the corresponding Cartesian axes. In spherical polar co-ordinates this becomes:

$$\frac{\gamma\left(\theta,\phi\right)}{\gamma_{0}} = (1-3\epsilon_{4}) \left[1 - \frac{4\epsilon_{4}}{(1-3\epsilon_{4})} (\sin^{4}\theta \left(\sin^{4}\phi + \cos^{4}\phi\right) + \cos^{4}\theta) \right]$$
(4.14)

A 2-dimensional representation of this can be found by setting:

$$N_z = 0 \to \theta = \frac{\pi}{2} \tag{4.15}$$

giving:

$$\frac{\gamma(\phi)}{\gamma_0} = (1 - 3\epsilon_4) \left[1 - \frac{4\epsilon_4}{(1 - 3\epsilon_4)} (\sin^4 \phi + \cos^4 \phi) \right]$$
(4.16)
= $(1 - 3\epsilon_4) \left[1 - \frac{4\epsilon_4}{(1 - 3\epsilon_4)} (\frac{3}{4} + \frac{1}{4}\cos 4\phi) \right]$
= $1 + \epsilon_4 \cos 4\phi$ (4.17)

The surface stiffness can be expressed in terms of the Herring condition as:

$$\kappa\Gamma(\theta,\phi) = \frac{1}{\gamma_0} \sum_{i=1}^2 \left(\kappa_i \gamma(\theta,\phi) + \frac{\delta^2 \gamma(\theta,\phi)}{\delta \zeta_i^2} \right)$$
(4.18)

where κ_i are the principal curvatures and ζ_i are the angles along the principal directions. In 2-dimensions, where through the assumption the solidification front is infinite along the z-direction, indicates that one of the principal directions must always lie in the z-direction and has zero curvature, the other principal direction

must be orthogonal to this therefore and lies in the x-y plane, thus this condition leads to:

$$\Gamma(\phi) = \frac{1}{\gamma_0} \left(\gamma(\phi) + \frac{\delta^2 \gamma(\phi)}{\delta \phi^2} \right)$$

= 1 - 15\epsilon_4 \cos 4\phi (4.19)

A recent proposal from the work by Qin *et. al* [97] using principels from magnetocrystalline anisotropy and the embedded-atom method (EAM) gives the form to be:

$$\gamma(\theta, \phi) = \alpha_0 + \alpha_1 (N_x^2 N_y^2 + N_y^2 N_z^2 + N_x^2 N_z^2) + \alpha_2 N_x^2 N_y^2 N_z^2 + \alpha_3 (N_x^2 N_y^2 + N_y^2 N_z^2 + N_x^2 N_z^2)^2 + \dots \gamma(\theta, \phi) = \alpha_0 + \alpha_1 (\sin^4 \theta \sin^2 \phi \cos^2 \phi + \sin^2 \theta \cos^2 \theta) + \alpha_2 (\sin^4 \theta \cos^2 \theta \sin^2 \phi \cos^2 \phi)$$
(4.20)

where for Aluminium $\alpha_0 = 0.922$, $\alpha_1 = 1.363$ and $\alpha_2 = -5.691$. Although the harmonics are not orthogonal they still exhibit cubic symmetry and the authors give plausible values for the anisotropic coefficients for real materials. Although an in depth study is necessary to see the consequence of the different formulations it is beyond the scope of this thesis, however it interesting to note that:

$$\sin^4\theta \left(\sin^4\phi + \cos^4\phi\right) + \cos^4\theta = 1 - 2(\sin^4\theta \sin^2\phi \cos^2\phi + \sin^2\theta \cos^2\theta) \quad (4.21)$$

highlighting the link between the two.

For the 3-dimensional surface stiffness, it is assumed that it follows the same cubic harmonics as the surface energy and that the surface stiffness can be related to the 2-dimensional version in equation 4.19 by assuming that the principal curvatures act in the same directions at the tip and that the surface stiffness is equivalent. This allows values of α_0 and α_1 to be related to ϵ_4 in 3-dimensions. However since the third harmonic has no physical meaning in 2-dimensions $\alpha_2 = 0$ is assumed for this comparison, figure 4.1 shows plots of the surface energy for both cases.



Figure 4.1: Surface energy of Aluminium. Top: Including terms up to α_2 , Bottom: Including terms up to α_1

Selection of the cell sized plays a crucial role in producing an accurate result. If the cell size is too large then the finite difference approximations for estimating the local curvature and normal vector become erroneous. Grid refinement is normally considered to improve the accuracy of a result, however by the definition of the enthalpy method where only a single cell is used to represent the interface, the interface thickness which is not directly specified scales proportionately with the cell size and so at some point refinement will provide no added benefit to the result. This is known as the 'Narrow Band Limit' [98] and refinement past this point is essentially a waste of computational power and may actually reduce the accuracy. The optimal cell size is therefore in the region where the cell size is small enough to accurately estimate the finite difference terms and large enough to extend the domain to capture the evolution of the moving front for a long period of time. The implementation of the enthalpy method in this work is directly related to Voller [31], where a quantitative study of mesh dependence and the maximum width of a cell before the accuracy begins to deteriorate was around $\Delta x = 5$. This value is used consistently throughout this thesis for both 2-dimensional and 3-dimensional cases; although a similar study is yet to be conducted in 3dimensions, a qualitative comparison is conducted at the beginning of the results section.

4.4 Potential Solver

The solution to the net potential is given from equations 2.7. If surface charge is assumed not to be present then the electric potential term can be neglected and the boundary can be written as:

$$\Psi_s^i = -\Psi_l^i \tag{4.22}$$

This implies a discontinuity at the interface, however by finding the solution to $-\Psi_l$ then only 1 boundary condition needs to be applied for both sides. From the formulation of the domain decomposition the boundary should be placed

at f = 0.5, which will exist in the interfacial cells, however the exact location inside these cells is not clearly defined. Thus to improve the accuracy a sub-mesh is implemented on cells close to the interface. A distance function calculates which cells should be sub-meshed by taking a locus of the interface for a given distance and then populating cells within this locus with a uniform Cartesian odd number of cells. The choice of an odd number of cells simply means that in terms of the finite difference solution the central cell of the sub-mesh will correspond to the cell centre of the macro mesh. Interpolated values for f and T are then populated in the sub-meshed cells. For interfacial cells this provides a more accurate representation of the step function approximation; allowing a tighter criteria to be used to distinguish between the liquid and solid, providing a better approximation for the interfacial potential boundary condition. The distance function is also used to model infinity; after a certain distance cells are tagged and the boundary condition at infinity is placed there. Cells beyond this are unsolved and \mathbf{J} is assumed to be zero, a representation of this is given in figure 4.2. Once the macro cells are tagged as either unsolved, infinity boundary, solved or sub-meshed, the solution procedure begins. The method is analogous to a multi-grid method and in this case has 3 steps. The first begins by imposing the infinity boundary, which is essentially an average value of the neighbouring solved cells:

$$\Psi_{\infty} = \sum_{i=0}^{n_s} \frac{\Psi_i}{n_s} \tag{4.23}$$

where n represents the solved neighbours and n_s is the total number of solved neighbours. The second performs an iteration on the sub-mesh by:

$$\Psi_{p} = \begin{cases} \sum_{i=0}^{n_{s}} \frac{\Psi_{i}}{n_{s}} & f < 0.5 \left(1 - \frac{1}{n_{sub}}\right) \\ \frac{1}{2} \Delta S & 0.5 \left(1 - \frac{1}{n_{sub}}\right) < f < 0.5 \left(1 + \frac{1}{n_{sub}}\right) \\ \sum_{i=0}^{n_{s}} \frac{\Psi_{i}}{n_{s}} & f > 0.5 \left(1 + \frac{1}{n_{sub}}\right) \end{cases}$$
(4.24)

where n_{sub} is the number of sub-mesh cells in each direction and the associated extra term controls the criteria for distinguishing between the liquid and solid and is sufficiently large to ensure that the interface is captured between two adjacent macro cells. The value at the central sub-mesh cell is passed to the corresponding macro mesh cell and an iteration is performed on the macro mesh, which is identical to 4.24 except that there will be no intermediate values of f:

$$\Psi_{p} = \begin{cases} \sum_{i=0}^{n_{s}} \frac{\Psi_{i}}{n_{s}} & f = 0\\ \sum_{i=0}^{n_{s}} \frac{\Psi_{i}}{n_{s}} & f = 1 \end{cases}$$
(4.25)

Finally the solution to macro cells that exist next to sub-meshed cells is used to provide a boundary condition for sub-meshed cells and iterations are then performed on the sub-mesh. This continues until convergence of Ψ . The current density can be calculated by taking the gradient in terms of finite differences on the macro and sub mesh. The normalisation of f, accounts for the solution of
$-\Psi_l$:

$$\mathbf{J} = \frac{-\frac{f(i,j,k)-0.5}{|f(i,j,k)-0.5|} \left(\Psi(i+1,j,k) - \Psi(i,j,k)\right)}{dx} \hat{x} \\ -\frac{\frac{f(i,j,k)-0.5}{|f(i,j,k)-0.5|} \left(\Psi(i,j+1,k) - \Psi(i,j,k)\right)}{dx} \hat{y} \\ -\frac{\frac{f(i,j,k)-0.5}{|f(i,j,k)-0.5|} \left(\Psi(i,j,k+1) - \Psi(i,j,k)\right)}{dx} \hat{z}$$
(4.26)

The average value of the sub-mesh can then be used to calculate its corresponding macro cell value. Finally by calculating $\mathbf{J} \times \mathbf{B}$ the Lorentz force can be passed to the flow solver. The sub-mesh will provide a more accurate calculation of the direction and magnitude of \mathbf{J} especially if the magnitude rapidly decays from the interface.

The locus method essentially counts cells away from the interface, by sweeping across the domain for each increment of a cell. The advantage of calculating the distance in this fashion does not restrict the domain from having more than one crystal. As two or more crystals begin to interact then the loci of all the crystals will form a single locus around all the crystals.

To demonstrate the increased accuracy of this method consider a 1-dimensional cross section normal to the solid-liquid interface. A representation of this is given in figure 4.3, where for this hypothetical example the macro-mesh cell centres lie on integer values of x, the sub-mesh contains $n_{sub} = 5$ cells per macro-mesh cell, the interface cell is located at x = 4 and is given the value f = 0.9. With only a single interface cell used to represent the interface it is necessary to assume that the temperature and liquid fraction at the cell centre are exact and that they vary linearly between the liquid and solid neighbours. Thus the sub-mesh is populated with values for the liquid fraction and temperature by linear interpolation



Figure 4.2: The Locus function calculates the type of solver to use for each cell.

from the macro-mesh cells (2 neighbours in 1-dimension, 4 in 2-dimensions and 8 in 3-dimensions). For the temperature it is also possible to solve the steady state heat equation using the macro-mesh cells as boundary conditions for the central sub-mesh cell. In this 1-dimensional case both methods are linear and identical, however for higher dimensions solving the heat equation provides small influences from cells that are not used in the local linear interpolation and may provide a small and perhaps more accurate modification. Using the constraint $0.5\left(1-\frac{1}{n_{sub}}\right) < f < 0.5\left(1+\frac{1}{n_{sub}}\right)$ to determine the interface guarantees that the interface will be captured. Figure 4.4 shows for this particular case that the location and temperature value used for the potential boundary condition become significantly more accurate between the macro-mesh interface cell and the submesh interface cell. In situations where $f \sim 0.5$ in the macro-mesh interface cell this technique will not provide any increase in accuracy in determining the location of the interface as it is already known. However in higher dimensions when the interface becomes a collection of cells with a variety of liquid fraction values it becomes necessary to implement such a technique to improve the accuracy.

4.5 Computational Fluid Dynamics

The solution to Navier-Stokes equations is given through a cell centred finite volume solver. The solver PHYSICA was developed by Croft *et al.* [99]. The solver uses the SIMPLEC method which follows the idea of a pressure correction method originally proposed by Patankar *et al.* [100] and later enhanced by Van Doormal *et al.* [101]. The exact derivation is not given here, however the final form for each of the discretised terms of Naiver-Stokes equation integrated in time



Figure 4.3: Hypothetical 1-dimensional cross section normal to the interface.



Figure 4.4: Hypothetical solutions for the location of the interface using the macro-mesh cell centre and the sub-mesh cell centre

and space are given below. From the Theory section Navier-Stokes equations are defined as:

$$\frac{\partial \rho \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u} = -\nabla p + \mu \nabla^2 \mathbf{u} + \mathbf{J} \times \mathbf{B} - K(f)$$
(4.27)

Integrating the transient term over time and a control volume leads to:

$$\int_{t-\Delta t}^{t} \int_{V} \frac{\partial(\rho\varphi)}{\partial t} dV dt \sim \frac{V_{p}^{t+1}\rho_{p}^{t+1}\varphi_{p}^{t+1} - V_{p}^{t}\rho_{p}^{t}\varphi_{p}^{t}}{\Delta t}$$
(4.28)

The conserved variable φ in this study represents the three components of velocity. The diffusion term becomes:

$$\int_{V} \nabla \cdot (\mu \nabla \varphi) dV = \int_{S} \mu \nabla \varphi \cdot \mathbf{n} dS \sim \sum_{f} \mu_{f} A_{f} \left(\frac{\varphi_{a} - \varphi_{p}}{d_{ap}} \right)$$
(4.29)

a represents the adjacent cell centre to p, A_f is the cross-sectional area of the face and d_{ap} is the distance between a and p. The convective term becomes:

$$\int_{V} \nabla \cdot (\rho \mathbf{u} \varphi) dV = \int_{S} \rho(\mathbf{u} \cdot \mathbf{n}) \varphi dS \sim \sum_{f} \rho_{f}(\mathbf{u} \cdot \mathbf{n})_{f} A_{f} \varphi_{f}$$
(4.30)

The Rhie - Chow interpolation method [102] is used to evaluate the term $(\mathbf{u} \cdot \mathbf{n})_f$ at the faces. The Lorentz force from the solution of the electric potential at cell centre is assumed to act uniformly over a control volume:

$$\int_{V} \mathbf{J} \cdot \mathbf{B} dV = (\mathbf{J} \cdot \mathbf{B})V \tag{4.31}$$

The final term is a Darcy resistance term which prevents flow from entering solidified regions and is defined by:

$$K(f) = \begin{cases} K & f < 0.5 \\ 0 & f > 0.5 \end{cases}$$
(4.32)

where K on the right hand side is significantly large driving the velocity field inside the solid to zero. These equations are solved using a Hybrid differencing scheme [103] and a conjugate gradient solver. For numerical stability relaxation parameters are used for the solutions of the pressure and momentum. For the pressure a linear relaxation is applied given by:

$$\varphi_{new} = \varphi_{old} + \upsilon(\varphi_{new} - \varphi_{old}) \tag{4.33}$$

where $0 < v \leq 1$ is the linear relaxation parameter and the new and old subscripts represent values between consecutive iterative sweeps. A momentum false time step is used in the solution for the momentum. It is based on the Courant Friedrichs Lewy condition [104] and takes the form:

$$dt_{false} < \min\left(\frac{\Delta x}{|\mathbf{u}|}\right) \tag{4.34}$$

which represents the minimum time taken for flow to pass through a single cell. The false time step essentially limits the change in momentum during the solution of a real time step by adding a pseudo-transient term to the solved conservation equation.

4.6 Initialisation

The domain is populated with initial values for all solved variables that are representative of a meta-stable state prior to nucleation. The domain is assumed to be fully liquid f = 1, undercooled and of a uniform concentration. For some cases an initial velocity field is also present.

4.6.1 Nucleation

At some time t = 0, a spherical seed nucleates in the centre of the domain. The seed is assumed to have a radius of 2dx and to represent this in terms of a liquid fraction it is necessary to calculate the volumetric proportions of the intersection of a circle and a square. This can be solved analytically by first noting that any cell which has the distance between its vertices and the origin less than the radius of the seed will be completely solid f = 0 and conversely if all of the vertices exist outside of the seed then it will be fully liquid f = 1. For this specific case only the cell area occupied by the hashed cell in figure 4.5 needs to be found in order to calculate the volumetric proportions of the rest of the cells. Rearranging the



Figure 4.5: Calculating the volumetric proportions of a square and a circle is used to calculate the initial liquid fraction values of a seed

equation of a circle:

$$y = (r^2 - x^2)^{\frac{1}{2}} \tag{4.35}$$

and integrating between 0 and dx and taking into account the area of the lower left cell gives:

$$\int_{0}^{dx} (r^{2} - x^{2})^{\frac{1}{2}} dx = \frac{1}{2} \left[\tan^{-1} \left(\frac{x}{(r^{2} - x^{2})^{\frac{1}{2}}} \right) r^{2} + x(r^{2} - x^{2})^{\frac{1}{2}} \right]_{0}^{dx} - 1 \quad (4.36)$$

which represents the hashed area. By symmetry the lower right cell will have the same value. The upper right cell is then calculated by:

$$A_{i+1,j+1} = \frac{\pi r^2}{4} - 1 - 2\int_0^{dx} (r^2 - x^2)^{\frac{1}{2}} dx$$
(4.37)

Finally $f_{i,j} = 1 - A_{i,j}$ gives the volumetric proportion for a quarter of the circle, the other quadrants can be calculated by symmetry. In 3-dimensions an analytic solution to the intersection of a sphere and cube may exist, but becomes rather complicated, thus a numerical scheme is used to calculate the volumetric proportions. The algorithm is a recursive algorithm that successively divides the cube into small cubes testing each of the vertices and adding the contribution to the volumetric proportions. A typical seed in the positive x, y, z quadrant of the domain has the following values in 2-dimensions:

0.087	0.685
0.000	0.087

In 3-dimensions:

$$\begin{array}{ccc} 0.183 & 0.764 \\ 0.000 & 0.183 \end{array}$$

and in the k + 1 plane:

0.764	0.984
0.183	0.764

Equations 4.9 and 4.10 are used to calculate corresponding values of T and C representative of the initial liquid fraction of the seed.



Figure 4.6: Recursive algorithms to calculate volumetric proportion of seed in 3D

4.7 Quasi 3-Dimensional Approximation

This section develops an approximation for some simulations carried out in the x-y plane in 2D. For any solved variable A, classically it is permitted to use approximations that $\frac{dA}{dz} = 0$ and w = 0, for cases exploring the effects of a magnetic field it can be shown that this is a highly erroneous and leads to a stagnant flow. Consider a section of incompressible fluid with initially stagnant flow with a conserved current circulation in the x-y plane under the influence of an external magnetic field B_z , this leads to a Lorentz force:

$$\mathbf{J} \times \mathbf{B} = \begin{vmatrix} i & j & k \\ J_x & J_y & 0 \\ 0 & 0 & B_z \end{vmatrix} = J_y B_z \hat{i}, -J_x B_z \hat{j}$$
(4.38)

acting only in the x-y plane as shown in figure 4.7. Taking the Curl of Navier-Stokes equation 2.13 gives the Vorticity equation:



Figure 4.7: Direction of Lorentz forces for a circulating current

$$\frac{\partial\omega}{\partial t} + \mathbf{u} \cdot \nabla\omega - \omega \cdot \nabla\mathbf{u} = \mu \nabla^2 \omega + \nabla \times \mathbf{J} \times \mathbf{B}$$
(4.39)

where:

$$\omega = \nabla \times \mathbf{u} = \left(\frac{\partial w}{\partial y} - \frac{\partial v}{\partial z}\right)\hat{i}, \left(\frac{\partial u}{\partial z} - \frac{\partial w}{\partial x}\right)\hat{j}, \left(\frac{\partial v}{\partial x} - \frac{\partial u}{\partial y}\right)\hat{k}$$
(4.40)

the final term is given by:

$$\nabla \times \mathbf{J} \times \mathbf{B} = \begin{vmatrix} i & j & k \\ \frac{\partial}{\partial x} & \frac{\partial}{\partial y} & \frac{\partial}{\partial z} \\ J_y B_z & -J_x B_z & 0 \end{vmatrix} = \frac{\partial J_x B_z}{\partial z} \hat{i}, -\frac{\partial J_y B_z}{\partial z} \hat{j}, -\left(\frac{\partial J_x B_z}{\partial x} + \frac{\partial J_y B_z}{\partial y}\right) \hat{k}$$
(4.41)

From the assumption that the flow is initially stagnant the convective, the diffusion and the stretching of vorticity terms become zero, so that any initial acceleration is purely from the external Lorentz forces leads to the following set of equations:

$$\frac{\partial}{\partial t} \left(\frac{\partial w}{\partial y} - \frac{\partial v}{\partial z} \right) = \frac{\partial J_x B_z}{\partial z} \tag{4.42}$$

$$\frac{\partial}{\partial t} \left(\frac{\partial u}{\partial z} - \frac{\partial w}{\partial x} \right) = -\frac{\partial J_y B_z}{\partial z} \tag{4.43}$$

$$\frac{\partial}{\partial t} \left(\frac{\partial v}{\partial x} - \frac{\partial u}{\partial y} \right) = - \left(\frac{\partial J_x B_z}{\partial x} + \frac{\partial J_y B_z}{\partial y} \right) \tag{4.44}$$

Applying the 2-dimensional approximations causes the first two equations to become 0. The right hand side of the final equation is equivalent to 2-dimensional continuity of **J**:

$$B_z \nabla \cdot \mathbf{J} = 0 \tag{4.45}$$

thus:

$$\frac{\partial\omega}{\partial t} = 0 \tag{4.46}$$

This implies that if a fluid particle has no initial vorticity it may never acquire it. It is then possible to define a scalar potential φ_u as:

$$\mathbf{u} = \nabla \varphi_u \tag{4.47}$$

from conservation of mass this leads to:

$$\nabla \cdot \mathbf{u} = \nabla^2 \varphi_u = 0 \tag{4.48}$$

The solution to φ_u satisfies Laplace's equation with far field boundary conditions of $\mathbf{u} = 0$. Thus the trivial solution $\varphi_u = 0$ and $\mathbf{u} = 0$ across the whole domain.

Introduction of the solid-liquid interface follows a similar argument; except half of the Lorentz forces are balanced by introducing some mechanical body force due to atomic bonding that is equal and opposite to the Lorentz force inside the solid as any net force would cause an acceleration. The force balance on the liquid side will manifest as a pressure gradient normal to the interface and equal and opposite to the Lorentz force. Applying the boundary condition at the interface $\mathbf{u} = 0$ still yields a stagnant flow solution to Laplace's equation. In 3 dimensional space this is not the case and can be highlighted in 4.44 where the right hand side is not necessarily zero by continuity. It is therefore necessary to change the 2D assumptions to account for forces of this nature.

The assumption $\frac{\partial f}{\partial z} = 0$, assumes that the solid is infinitely long in the zdirection and is the cause of the stagnant flow solution. Defining a height function f(z), which accounts for the 3D morphology of the crystal implies that the pressure gradient normal to the interface above and below the solid (i.e. in the zdirection) will in general not be equivalent to the Lorentz forces in the x-y plane. The simplest height function is to assume that the crystal has zero thickness in the z-direction:

$$f(z) = \begin{cases} 1 & z < 0 \\ 0 & z = 0 \\ 1 & z > 0 \end{cases}$$
(4.49)

Selecting this height function causes the normal to the interface to exist only in the z-direction and therefore there are no contributions to the pressure gradient in the x-y plane to balance the Lorentz forces in the liquid. This effectively allows fluid flow to pass above and below the crystal without having to directly model the changes in w, which through symmetry can still be considered w = 0. This is implemented by simply removing the resistive term in 2.13 and flow appears to penetrate into the solid regions of the crystal. With no knowledge of the real crystal morphology and the interface temperature in the z-direction, the thermoelectric currents can only be solved in the x-y plane, therefore it is assumed that the Lorentz forces calculated in this region can be applied in planes where $z \neq 0$. Values for the transported variables in the solid are not representative of the liquid phase, to take into account the transport equations as flow passes around the crystal it is assumed that the flow acts purely in the boundary layer and a second set of scalar variables Tz and Cz are introduced and initialised with typical values representative of the boundary layer. As flow passes around the crystal the convective transport is calculated from these variables instead. Consider a boundary between a liquid cell and a solid cell, the transport equations at the liquid cell are modified to:

$$H_{i}^{t+1} = H_{i}^{t} + \frac{\Delta t}{\Delta x} \left(\left(\frac{T_{i-1}^{t} - T_{i}^{t}}{\Delta x} \right) + \frac{\left(T_{i-1}^{t} + T_{i}^{t}\right) \left(u_{i-1}^{t} + u_{i}^{t}\right)}{4} - \left(\frac{T_{i}^{t} - T_{i+1}^{t}}{\Delta x} \right) + \frac{\left(T_{i}^{t} + Tz_{i+1}^{t}\right) \left(u_{i}^{t} + u_{i+1}^{t}\right)}{4} \right)$$
(4.50)

a 1-dimensional representation of this is given in figure 4.8. The transport of Hz is unmodified as the cell is still liquid and the only difference is the convective flux out of the cell which uses the the value of Tz. In the solid side values of H and Hz are updated by:

$$H_{i}^{t+1} = H_{i}^{t} + \frac{\Delta t}{\Delta x^{2}} \left(T_{i-1}^{t} - 2T_{i}^{t} + T_{i+1}^{t} \right)$$

$$Hz_{i}^{t+1} = Hz_{i}^{t} + \frac{\Delta t}{\Delta x} \left(\left(\frac{\left(T_{i-1}^{t} + Tz_{i}^{t} \right) \left(u_{i-1}^{t} + u_{i}^{t} \right)}{4} \right) - \left(\frac{\left(Tz_{i}^{t} + Tz_{i+1}^{t} \right) \left(u_{i}^{t} + u_{i+1}^{t} \right)}{4} \right) \right)$$

$$(4.51)$$

$$(4.52)$$

where a representation of this is given in figure 4.9.



Figure 4.8: Discretisation of the transport equations for the quasi 3-dimensional approximation on the liquid side of the interface



Figure 4.9: Discretisation of the transport equations for the quasi 3-dimensional approximation on the solid side of the interface

4.8 Moving Mesh

For certain aspects of this research the concept of a moving mesh is introduced. The motivation for this technique is to perform a detailed analysis of time independent solutions to compare to analytic solutions, where a conventional mesh is too small and far field boundary condition approximations are no longer valid. For the moving mesh to be valid it is assumed that historical data moving out of the mesh no longer have an influence on the overall solution. The implementation of this keeps track of the dendrite tip and the mesh moves such that the tip is always central to the domain, this essentially gives a solution in the moving mesh is tracking a tip growing in the positive x-direction, when the tip grows by a single cell the mesh will shift and all time-dependent variables $(A_{i,j,k}^t)$ will be moved by:

$$A_{i,j,k}^t = A_{i+1,j,k}^t \tag{4.53}$$

This applies everywhere in the domain except the last row of cells at i = nx, where it is necessary to keep a meaningful boundary condition. For Dirichlet boundaries:

$$A_{nx,j,k}^t = C \tag{4.54}$$

and for Von-Neumann, the layer of unsolved but updated boundary cells at (nx + 1) is used:

$$A_{nx,j,k}^{t} = A_{nx+1,j,k}^{t} \tag{4.55}$$

For the face where data are being removed, the following boundary conditions are applied:

$$\frac{\partial f}{\partial \hat{n}} = \frac{\partial T}{\partial \hat{n}} = \frac{\partial C}{\partial \hat{n}} = \frac{\partial \Psi}{\partial \hat{n}} = 0 \tag{4.56}$$

p =

$$= 0$$
 (4.57)

4.9 Complete Algorithm

The full algorithm is given in figure 4.10. The largest proportion of computational time is spent solving the fluid dynamics, in general there is a disparity in the required time step size between the solidification solver (t_s) and the fluid dynamics solver (t_f) . Therefore a sub-stepping technique is introduced to speed up the time taken to perform the total simulation. The step size is chosen such that the interface cannot grow further than a single cell. The most significant reason for this choice is related to circulations that form at the tip in cases involving an external magnetic field. Given that the smallest diameter that a circulation can exist numerically is two cells as the velocity can only have a single direction in a cell. As will be discussed later the Lorentz force re-accelerates this circulation as the interface moves thus if t_f is too large the interface will have moved through the circulation and the corresponding impulse from the Lorentz force will be over predicted.

After initialisation, the transport equations evolve the temperature and solute fields and the seed grows through the solidification algorithm. When $t_s = dt_f$, the sub-mesh is set up for this particular instantaneous location of the solid front, the electric potential is solved and the Lorentz force is calculated. The Lorentz force and liquid fraction are given to the flow solver and the change in the velocity field is computed. The flow solver returns a new value for the velocity which then modifies the transport equations changing the evolution of the crystal morphology.



Figure 4.10: Flow diagram of the complete algorithm

4.10 Summary

The numerical algorithms used in this research are outlined in this chapter. A set of dimensionless scaling factors are introduced and each of the three solvers (soldification, thermoelectricity and magnetohydrodynamics) are detailed explicitly in a discretised form. The algorithms for nucleation, a quasi 3-dimensional approximation and a moving mesh technique are also described. Finally for fully coupled simulations the complete algorithm is given.

Chapter 5

Results

5.1 Overview

This chapter explores the implementation of the numerical model and the predictions that are made. Several cases are investigated each introducing an increased detail of the physical phenomena and for each case simulations are presented in both 2-dimensions and 3-dimensions. The first test looks at a pure material with transport through diffusion, the second case introduces a directional forced convection, the third case investigates the flow patterns that emerge when an external magnetic field is applied and the fully coupled cases investigate the effects of thermoelectric magnetohydrodynamics in both a pure material and a binary alloy.

5.2 Diffusion Driven Growth

5.2.1 Effect of Anisotropic Strength on Crystal Morphology

The effect of anisotropy on crystal morphology is investigated by holding the other key parameters constant. The material is assumed to be pure, the initial undercooling is set to T = -0.5 and $\mathbf{u} = 0$. For a spherical seed with no anisotropic strength growing with no random fluctuations then the contributions to the growth should be equivalent in all directions as κ is constant at every position on the interface. However this solution is an unstable equilibrium and any small perturbation will deform the spherical growth [6]. In nature thermal and solute fluctuations cause the sphere to destabilise, numerically these fluctuations occur through mesh errors, rounding errors and can also be intentionally imposed. For the purpose of this study the latter is not included and the calculations are performed to at least 8 significant figures making rounding errors very small such that it will take a long time to observe the effect on morphological stability of a sphere. The largest error is therefore due to the mesh, where modelling a sphere on a Cartesian grid inherently introduces errors. Quantifying the influence of this error directly is not straight forward as an initial small perturbation will ultimately grow unbounded. To explore these implications and give some appreciation to the direction of this error the anisotropic strength is set to 0 i.e. $\gamma(\theta, \phi) = 1$. Figure 5.1 shows the 2D results at two separate times, the first at t = 2500 shows how small errors are beginning to deform the circular shape and the second at t = 250000 shows the end result. The mesh essentially introduces anisotropy and in this case causing the crystal to grow preferentially in the (100)

and (110) directions. This preference is not unexpected as it is in the direction of the computational neighbours.

In 3-dimensions a similar situation is observed; at an early stage of growth given in figure 5.2, the crystal grows somewhat spherical until perturbations of the interface form. The results at a much later stage are given in figure 5.3, where these perturbations, which have continued to grow unbounded giving similar morphological changes to 2 dimensions. However with the extra degree of freedom included there is also a preference in the (111) direction.

Introducing a small amount of anisotropy to represent an equiaxed crystal lattice, the spherical shape is immediately deformed and for equiaxed crystals the preferential growth direction becomes purely the (100) direction. The effect on the crystal morphology as ϵ_4 is increased is shown in figure 5.4 as the anisotropic strength increases the tip radius decreases and the arms become more needle like. This trend is expected as the anisotropic term decreases the free energy required for solidification to occur in the (100) direction and increases the free energy required in the (110) direction.

In 3-dimensions; using an equivalent $\epsilon_4 = 0.05$ the crystal morphology is given in figure 5.5. The dendrite grows in the (001) direction and exhibits similar morphological features to the 2-dimensional case and also qualitatively agrees with the results presented in figure 3.2 and 3.3 in the review section [32].

The 3rd harmonic from equation 4.20 has no physical representation in 2dimensions, as the assumption $N_z = 0$ prevents this. However in the 3-dimensional simulation it can be included and the results are given in figure 5.6 and although the crystal still exhibits an equiaxed structure the (111) direction is also taken into consideration.



Figure 5.1: 2-D simulation of curvature driven growth showing the crystal morphology and thermal field at two stages of growth. Top: t = 2500, Bottom: t = 250000



Figure 5.2: 3-D simulation of curvature driven growth showing the crystal morphology and thermal field at t = 3000



Figure 5.3: 3-D simulation of curvature driven growth showing the crystal morphology and thermal field at t = 9000



Figure 5.4: 2D growth of a pure material at t = 50000 with various values for ϵ_4 . Top left: $\epsilon_4 = 0.01$, Top right: $\epsilon_4 = 0.02$, Bottom left: $\epsilon_4 = 0.03$, Bottom right: $\epsilon_4 = 0.05$



Figure 5.5: 3D growth of a pure material at t = 30000 with $\epsilon_4 = 0.05$



Figure 5.6: 3D growth of a pure Aluminium using the first 3 harmonics at t = 40000

Removing the 3rd harmonic provides the best representation of how the 2dimensional model corresponds to the 3-dimensional model. The result when only the first 2 harmonics are included is given in figure 5.7, the tip morphology and velocity are very similar to the cases with 3 harmonics, however the tip growing in the (111) direction is now missing. This highlights two important features; the first is that the the third harmonic does not affect the 2-dimensional simulations since the tip velocity is unchanged. The second is that the anisotropy is controlling the growth and not the numerical error through the mesh that is seen in the case where the dendrite is curvature driven only and the surface energy is isotropic. The curvature driven case has preference in the (001), (011) and (111) directions, which is analogous to the case where all three cubic harmonics are considered for Aluminium. The absence of the (111) branch when the third harmonic is removed shows that the surface energy anisotropy is the dominating factor and the crystal morphology is not controlled by the mesh, for the cell size used.

From microscopic solvability theory the tip velocity should approach a constant. Utilising the moving mesh technique the steady state tip velocity can be calculated as the influence from the neighbouring primary arms diminishes. It is not possible to relate directly α_0 and α_1 for Aluminium to some value of ϵ_4 , instead the steady state tip velocity for Aluminium is related to the corresponding value for ϵ_4 . Figure 5.8 shows the steady state tip velocity (u_{tip}) as a function of anisotropy for 2-dimensions and 3-dimensions, in both cases the trend is linear. Solving the point where the 3-dimensional linear trend intersects with the Aluminium tip velocity ultimately gives an estimate of $\epsilon_4^{Al} \sim 0.017$ that can be used to simulate Aluminium in 2-dimensions.



Figure 5.7: 3D growth of a pure Aluminium using the first 2 harmonics at t = 40000



Figure 5.8: Steady State tip velocities for 3-dimensional and 2-dimensional growth

There is a significant difference between the tip velocity in 2D and 3D, which comes about through the assumptions made when reducing the problem into 2-dimensions. Under steady state conditions the equilibrium at the tip gives:

$$u_{tip}R_{tip} = C \tag{5.1}$$

Assuming the constant on the right is related such that:

$$\frac{\partial f}{\partial t} \propto C \tag{5.2}$$

then from the definition of enthalpy:

$$\frac{\partial T}{\partial t} \propto C \tag{5.3}$$

$$\nabla^2 T \propto C \tag{5.4}$$

$$\frac{u_{tip(3D)}R_{3D}}{u_{tip(2D)}R_{3D}} \propto \frac{\nabla^2 T_{3D}}{\nabla^2 T_{2D}}$$
(5.5)

(5.6)

Given that the 2D model is a representation of the 3D model in the (r, ϕ) plane where $\frac{\partial f}{\partial z} = 0$, implies that one of the principal curvatures lies in the z-direction and is 0. Thus when comparing a cylinder to a sphere of equal radii, the curvature in 3D is twice as large as in 2D. Also assuming that $\frac{\partial T}{\partial x} = \frac{\partial T}{\partial y} = \frac{\partial T}{\partial z}$ in 3D, but in 2D $\frac{\partial T}{\partial z} = 0$ and the constants of proportionality are equal then:

$$\frac{u_{tip(3D)}}{2u_{tip(2D)}} = 1.5\tag{5.7}$$

Giving $\frac{V_{3D}}{V_{2D}} = 3$. The numerical predictions give a value varying between $3.7 < \frac{V_{3D}}{V_{2D}} < 4.0$, which is similar but slightly more than predicted. In this simple derivation it is assumed that the constant on the right which describes the operational state of the dendrite is equivalent between 2D and 3D. Furthermore this derivation only includes the change through transport and no analysis of stability is taken into consideration. Therefore it is likely that this discrepancy may be a consequence of either of these assumptions, however for the purpose of indicating that there should be a significant increase in the velocity between the 3-dimensional and 2-dimensional model this is sufficient.

These preliminary results demonstrate the behaviour of the dendritic model and provide a set of base morphologies that can be used to compare against later cases when forced convection and magnetic fields are applied as it is essentially the relative change that will have any quantitative meaning.

5.3 Effect of Forced Convection on Crystal Morphology

In the presence of flow the final term in the transport equations will begin to influence the distribution of the thermal and solute fields. Initially only considering the thermal field a simple test case is used to compare this work to that of other authors found in the literature review. A constant velocity boundary is placed on the north wall and the same velocity is initialised throughout the domain. A constant pressure boundary is placed on the south wall and the temperature is initialised as a uniform undercooling of T = -0.5. The solution begins in a steady state condition and upon nucleation the thermal field will react through solidification, diffusion and convection. In this set up the flow is directed normal to the direction of crystallographic orientation. Figure 5.9 shows the morphological changes and thermal fields for a variety of different flow cases in 2 dimensions. The velocity fields are very similar aside from the change in magnitude and so only the velocity plot in the case where $\mathbf{u} = 0.02$ is given in figure 5.10.

The north tip is extended, while the downstream south tip is stunted. The east and west tips are identical due to the symmetry plane that exists along the centre of the domain. The flow incident on the north tip lowers the temperature field which increases the local free energy encouraging the tip to grow, however as the flow passes around the dendrite inside the thermal boundary layer the flow transports warmer liquid and a hot region forms downstream decreasing the local free energy at the south tip and stunting growth. This can be seen as the thermal boundary layer downstream of the dendrite is extended, while at the incident tip becomes very narrow. In all cases a similar process is occurring and the degree of



Figure 5.9: 2D thermal fields in the presence of forced convection at different fluid velocities. Top left: $\mathbf{u} = 0.005$ at t = 50000, Top right: $\mathbf{u} = 0.01$ at t = 50000, Bottom left: $\mathbf{u} = 0.02$ at t = 40000, Bottom right: $\mathbf{u} = 0.03$ at t = 38000



Figure 5.10: 2D velocity field with $\mathbf{u}=0.02$ at the north face.

morphological change from the stagnant cases increases as the velocity increases. Figures 5.11, 5.12, 5.13 and 5.14 show the relative tip velocities for each tip as the flow velocity is varied. As the velocity increases the tip velocities vary when compared to the stagnant growth case.



Figure 5.11: Tip velocities for each tip with a forced convection of 0.005

An interesting feature is that each tip is independently forming a constant tip velocity, which is indicative that an equilibrium between the release of latent heat and the transport of heat is forming at each tip when flow is included. The steady state tip velocities for each tip as a function of the flow velocity are given in figure 5.15, which shows that as the velocity increases the south tip velocity becomes very small approaching zero while the other tips seem to increase in velocity when compared to the stagnant case.

The flow so far can be considered as potential flow, where the Reynolds number can be considered to be small the fluid essentially follows the solid-liquid interface. However under certain conditions, when the Reynolds number is high



Figure 5.12: Tip velocities for each tip with a forced convection of 0.01



Figure 5.13: Tip velocities for each tip with a forced convection of 0.02


Figure 5.14: Tip velocities for each tip with a forced convection of 0.03



Figure 5.15: Steady state tip velocity for each tip as in the presence of a forced convection

this is no longer the case. Quantifying the Reynolds number is not a straight forward process, as the characteristic length scale is not well defined. It is dependent on the size and morphology of the dendrite, which ultimately means that the Reynolds number is time dependent. Assuming that the length of the dendrite perpendicular to the flow is a valid approximation for the Reynolds number then upon nucleation the Reynolds number will be close to zero and potential flow will exist and the transport mechanism will be analogous to the results presented so far. As the dendrite grows and the length scale increases at some critical time circulations will form and the transport mechanism will change. However using a low anisotropic parameter with low tip velocities allows the extended down stream thermal field to form, when the circulations form it then takes a long time for the thermal field to redistribute itself. For this reason a high anisotropic $\epsilon_4 = 0.05$ is used and the flow velocity is increased to $\mathbf{u} = 0.2$ causing the transition of flow to occur faster. Figure 5.16 shows downstream circulations either side of the south tip, in a similar fashion to figure 3.8. These circulations prevent the thermal field from simply extending downstream of the dendrite and instead create a region where the southern tip can form, this can be seen in figure 5.17. The Reynolds number at the final time is approximately 1000, which exists in the region of laminar flow, but is unlikely to have transitioned into turbulent flow.

The same situation is set up in 3-dimensions, where the fundamental difference now is that flow is able to pass above and below the crystal and is not restricted to going around. The results are given in figures 5.18, 5.19 and 5.20 for the thermal field, direction of velocity and magnitude of velocity respectively. Similar morphological changes can be seen to the 2-dimensional case with low Reynold numbers; the incident tip is extended, the down stream tip is stunted and the



Figure 5.16: 2D velocity field with $\mathbf{u}=0.2$ at the north face at t=3400. $Re\sim1000$



Figure 5.17: 2D thermal field with a forced convection of $\mathbf{u} = 0.2$ at t = 3400. $Re \sim 1000$

perpendicular tips are all identical. However it is likely that much higher fluid velocities will be necessary to form any downstream circulations, as the increased degree of freedom for the fluid prevents this. Essentially the width of the dendrite is no longer a good approximation for the characteristic length and the radius of the crystal arm needs to be taken into consideration.

5.3.1 Growth with an Imposed Electric Potential in the Presence of a Magnetic Field

An interesting and somewhat similar situation to forced convection occurs when looking at imposed electrical potentials. Consider a material which is conducting when liquid and non-conducting when solid (for example an ionic compound). Thermoelectric currents therefore do not exist and when an artificial electric potential is placed across this material current will flow in the liquid from regions of high potential to low potential, where the non-conducting crystal can be represented as a change to the interface potential boundary condition:

$$\frac{\partial \Psi^i}{\partial \hat{n}} = 0 \tag{5.9}$$

A simple model was posed investigating a non-conducting dendrite nucleating under these conditions. Prior to nucleation and assuming that the imposed potential is time independent where the north face has a potential $\Psi = 1$ and the south face has a potential $\Psi = -1$ then the solution to the electric potential will be linear and consequently the current density will be constant throughout the domain. In the presence of a perpendicular magnetic field Lorentz forces will form throughout the material driving flow. In this particular case, with a positive magnetic field in the z-direction, a uniform force will form throughout



Figure 5.18: 3D thermal field with a forced convection of $\mathbf{u} = 0.02$ at t = 1600



Figure 5.19: Direction of velocity in 3D with $\mathbf{u}=0.02$ at the west face



Figure 5.20: Magnitude of velocity in 3D at a slice along z = 0

the domain in the negative x-direction. Assuming a steady state solution as the initial conditions prior to nucleation leads to a constant velocity in the negative x-direction. Upon nucleation the dendrite will grow influenced by the convective transport from the fluid dynamics, but also in turn changing the electric potential internal to the liquid; forcing current to pass around the dendrite due to the interfacial potential boundary condition. Figure 5.21 shows how the growing dendrite influences the electric potential and figure 5.22 shows the corresponding current density that forms; as the dendrite grows a region of high current forms close to the dendrite in the y-plane. However since the current density is now coupled to the dendrite morphology this in turn causes slight changes to the Lorentz forces close to the solid-liquid interface, the net effect provides a flow that is similar in direction to that of forced convection and is shown in figure 5.23. The surface energy anisotropy used is much higher than in the 3-dimensional forced convection case and so the morphological features although similar cannot be directly compared. However although the direction of velocity is similar, the magnitude of velocity is no longer symmetric at the tips perpendicular to the flow. The magnetic field removes part of the symmetry and the region of high current in the y-plane provides a higher Lorentz force at the x and z tips compared to the y tips. Consequently the fluid is faster in this region; this is highlighted in figure 5.24 which shows the magnitude of velocity.



Figure 5.21: Electric Potential of a 3D non-conducting dendrite with an imposed electric potential



Figure 5.22: Current density of a 3D non-conducting dendrite with an imposed electric potential



Figure 5.23: Direction of velocity of a 3D non-conducting dendrite with an imposed electric potential



Figure 5.24: Magnitude of velocity of a 3D non-conducting dendrite with an imposed electric potential

5.4 Direction of Flow Fields in the Presence of a Magnetic Field

Using the solution procedure with the boundary conditions detailed in section 4.4 the electric potential can be solved numerically on a growing dendrite. For simplicity assume that the melt is a pure material and that there is a significant Seebeck power between the solid and liquid. The case of a binary alloy will be considered later in this work, but to demonstrate the underlying principle driving the flow this assumption is sufficient. By taking a low magnetic field strength approximation the flow will be very small and the transport will be diffusion dominated. Any velocity field that forms will have no impact on the growth mechanics and the dendrite will grow identically to the case of diffusion driven growth in section 5.2. These assumptions essentially de-couple the growth equations from the convective transport, allowing for a simplified analysis of the flow field. The calculations in figure 5.7 were performed on a $200 \times 200 \times 200$ uniform Cartesian grid simulating only an octant of the problem by exploiting symmetry lines. However due to limitations on both memory and the computational time required to solve the problem when considering thermoelectric currents and Navier-stokes equations, the domain is reduced to $84 \times 84 \times 84$. Also to prevent the far field boundaries from influencing the solution of the electric potential and fluid flow the dendrite cannot be grown to the extremes of the domain. For these reasons the results presented are at a much earlier time and although the dendrite is not as well developed the morphological features are prominent. Estimates for the computational time required for all the cases presented in this work are given at the end of this chapter.

5.4 Direction of Flow Fields in the Presence of a Magnetic Field

For a given dendrite shape applying the sub-meshing locus gives a separate domain of cells that will be solved for the potential. Figure 5.25 shows the region of sub-meshed cells, solved cells, boundary cells and unsolved cells, this forms a shell around the dendrite avoiding the need to solve the electric potential throughout the domain, reducing the amount of computational time required.

Figure 5.26 shows the magnitude of the surface potential and a slice in the x-y plane of the far field potential. The unsolved cells have been populated with the average boundary cell value; there is no discernible difference between boundary cells and the unsolved cells indicating that the placement of the boundary condition is sufficiently far enough away to not have a significant influence on the solution. By taking the positive gradient of the potential on the liquid side (negative gradient on the solid side) the thermoelectric current density can be resolved. The direction of currents is given in figure 5.27, where the current travels from high potential regions to lower potential regions in the liquid and vice-versa in the solid forming a current circulation. The magnitude of the current density is given in figure 5.28. The 2D slice shows that the current density is localised very close to the interface; well within the thermal boundary layer, the current density reaches maxima at the tip of the dendrite and at the root.

For the 2-dimensional model the solution for the potential is given in figure 5.29 and the direction of J and magnitude are given in figure 5.30.

In both the 2-dimensional and 3-dimensional cases the current emanates from the tip of the dendrite and circulates between the root of the crystal. Physically this represents electron hole pairs splitting at the root of the crystal where due to the increased temperature electrons are more readily promoted to high energy levels, becoming more mobile and with a higher solute concentration there is a



Figure 5.25: Locus boundaries used in the submeshing technique. Top: 2D submesh along which is equivalent to the 3D sub-mesh along the z = 0 plane, Bottom: The corresponding 3-dimensional shell that forms



Figure 5.26: 3D potential in stagnant growth. Top: Slice at z = 0, Bottom: Surface potential



Figure 5.27: 3D direction of current density in stagnant growth



Figure 5.28: 3D magnitude of current density in stagnant growth. Top: Slice at z = 0, Bottom: Magnitude at the surface



Figure 5.29: 2D potential in stagnant growth



Figure 5.30: 2D direction and magnitude of current density in stagnant growth

larger difference in electron affinity. The electrons then travel to the tip of the crystal, falling into lower energy levels and recombining. The most significant difference between the 2D and 3D cases is that circulations of \mathbf{J} can only be formed in the *x-y* plane in 2D, while in 3D the current may take any path.

The formation of thermoelectric currents of this form exists for all equiaxed dendrites growing in an undercooled melt. However the corresponding Lorentz forces are dependent on the orientation of the magnetic field. When considering 2dimensional problems, the magnetic field must be aligned along the (001), (010)or (100) directions, such that the Lorentz forces will form in a 2D plane that satisfies the assumptions taken for 2-dimensional dendritic growth. For example for a magnetic field aligned in the z-direction (001) the Lorentz forces will exists purely in the x-y plane. When looking in 3-dimensional space where the magnetic field can be aligned in any direction and assuming a low magnetic field strength approximation the problem can be reduced into a small region of magnetic field orientations by exploiting the symmetry lines of \mathbf{J} . This is highlighted in figures 5.31 and 5.32, where the first set of symmetry lines are that of an octant i.e. planes perpendicular to the (001), (010) and (100) directions. A second set of symmetry lines also exist in the planes perpendicular to the $(\bar{1}10)$, $(10\bar{1})$ and $(0\bar{1}1)$ directions in the first octant. This reduces the problem to a region bounded by the (001), (011) and (111) directions and it is these extremes that will be considered as well as the (123) direction.

In 2 dimensions with the magnetic field directed in the (001) plane, such that $\mathbf{B} = (0, 0, B_z)$. Figure 5.33 shows the Lorentz forces acting in the *x-y* plane. In 3-dimensions the direction of the Lorentz force is given in figure 5.34 and the magnitude in figure 5.35. The 2-dimensional simulation essentially represents a



Figure 5.31: 3D representation of symmetry planes and the region of unique forces and flow fields



Figure 5.32: Ternary diagram illustrating the lines of symmetry and directions of magnetic field

slice through the x-y symmetry plane at z = 0. At this position the Lorentz forces emanate from a point at the interface and terminate at another point. However looking at the Lorentz forces just above the x, y arms a force exists in the anticlockwise direction; moving further along the z tip the force changes direction. The maximum force occurs in similar locations to the maximum J namely the tip and root.

Given that the co-ordinate system is chosen purely for numerical convenience and the dendrite will grow with symmetry planes regardless of the orientation of the co-ordinates chosen implies that aligning the magnetic field for example in the *y*-direction will generate Lorentz forces identical to the case where the magnetic field is aligned in the *z*-direction with a co-ordinate transformation. In this example this is represented by an Euler angle rotation with $\alpha = \frac{\pi}{2}$, $\beta = \frac{\pi}{2}$



Figure 5.33: 2D direction of Lorentz force in stagnant growth



Figure 5.34: Direction of Lorentz force with magnetic field directed in the (001) direction in different planes. Top left: Plane through the origin and perpendicular to the magnetic field, Top Right: Above x,y arms, Bottom left: Close to z tip, Bottom right: Isometric view



Figure 5.35: Magnitude of Lorentz force with magnetic field in (001) direction

and $\gamma = 0$ using the standard zxz transformation leading to:

$$\begin{bmatrix} x'\\ y'\\ z' \end{bmatrix} = \begin{bmatrix} 0 & 1 & 0\\ 0 & 0 & 1\\ 1 & 0 & 0 \end{bmatrix} \begin{bmatrix} x\\ y\\ z \end{bmatrix} = \begin{bmatrix} y\\ z\\ x \end{bmatrix}$$
(5.10)

where x', y' and z' represent the new axis. Applying this transformation to the Lorentz forces with the magnetic field directed in the z-direction would give the Lorentz forces with a magnetic field aligned along the y-direction. The same transformation can be applied again to give the Lorentz forces along the x-direction:

$$\begin{bmatrix} x'\\y'\\z' \end{bmatrix} = \begin{bmatrix} 0 & 0 & 1\\1 & 0 & 0\\0 & 1 & 0 \end{bmatrix} \begin{bmatrix} x\\y\\z \end{bmatrix} = \begin{bmatrix} z\\x\\y \end{bmatrix}$$
(5.11)

A consequence of the low magnetic field strength approximation is that the Lorentz force becomes linear and therefore the concept of linear superposition becomes valid. This allows the problem to be reduced even further; by creating a set of orthogonal basis functions, the force can be calculated for any orientation. The most obvious set is the Lorentz forces with the magnetic field in the (001), (010) and (100) directions, which represent the magnetic field aligned along the direction of crystallographic orientation. Physically irrespective of the tip chosen, as this is purely a matter of preference, the x, y and z tips can be interchanged through Euler angle rotations described previously. Thus it is only necessary to simulate the forces in a single direction to calculate the forces for any orientation.

The direction of the Lorentz forces are given in figure 5.36 and the relative magnitude in figure 5.37 when the magnetic field is orientated in the (011) direction. Aligning the magnetic field with the (011) direction gives rise to forces that are somewhat different to the (001) direction. When the magnetic field is orientated in the (011) direction similar force directions are observed in the plane

that passes through z = 0 as the (001) case, with the exception that the forces alternate in direction 4 times. Above the x arms the forces have the same direction as the incident forces on the interface as in the z = 0 plane and in the root between the y and z tips the forces form a closed loop.

With the magnetic field orientated in the (111) direction the direction of the forces are given in figure 5.38 and the relative magnitude in figure 5.39. In the plane that passes through z = 0 all the forces form a closed loop in the anticlockwise direction. Underneath the positive arms the forces are similar to the z = 0 plane in the (001), except the forces alternate in direction 6 times. Above the positive arms, the forces form closed loops around each of the tips and also in the root. Although the maxima occur in similar places in all of these cases the direction highlights how a variation in flow fields will also exist.

These extremes can be considered to be a secondary set of basis functions, providing an indication to the variety of forces that can be achieved by selecting the orientation of the magnetic field. Choosing an intermediate value such as the (123) direction features of all three of the secondary basis functions can be observed. This result is not unexpected as the forces just follow linear superposition, but the result does illustrate that selection of magnetic field orientation can play a critical role in the development of Lorentz forces.

Using the forces for each orientation of magnetic field and solving the Navier-Stokes equations yields the flow and pressure. The direction of the flow is given in figures 5.43, 5.45 and 5.47 for the (001), (011) and (111) directions respectively. The relative normalised velocities close to the surface of the dendrite are given in figures 5.44, 5.46 and 5.48 for the (001), (011) and (111) directions respectively. For the (001) case circulations form at the x-y tips, a global circulation forms



Figure 5.36: Direction of Lorentz force with magnetic field directed in the (011) direction in different planes. Top left: Plane through the origin and perpendicular to the magnetic field, Top Right: Above x arms, Bottom left: Between positive y and z arms, Bottom right: Isometric view



Figure 5.37: Magnitude of Lorentz force with magnetic field in (011) direction



Figure 5.38: Direction of Lorentz force with magnetic field directed in the (111) direction in different planes. Top left: Plane through the origin and perpendicular to the magnetic field, Top Right: Under positive x,y and z arms, Bottom left: Above positive x,y and z arms, Bottom right: Isometric view



Figure 5.39: Magnitude of Lorentz force with magnetic field in (111) direction



Figure 5.40: Direction of Lorentz force with magnetic field directed in the (123) direction in different planes. Top left: Plane through the origin and perpendicular to the magnetic field, Top Right: Above x arms, Bottom left: Above positive x,y and z arms, Bottom right: Isometric view



Figure 5.41: Magnitude of Lorentz force with magnetic field in (123) direction

just above the x-y arms in an anti-clockwise direction around the crystal and a circulation forms around the z-tip. When comparing this to the velocity field in 2-dimensions using the quasi 3-dimensional approximation, the features close to the z = 0 plane are quite well resolved, however the circulation around the z tip cannot be resolved in 2-dimensions, as the forces that cause this are not solved in 2-dimensions. This shows that the quasi 3-dimensional approximation is a necessity when considering forces of this nature. The maximum velocity is in the global circulation and the regions of high and low pressure alternate either side of the x-y tips.

In the (011) case circulations form similar to the (001) case, at the x-tips a global circulation passes over the x tips and under the z and y tips, circulations form at the z and y tips and a system of two circulations forms in the roots. Regions of high velocity are located in the global circulation and also around the circulations at the z and y tips.

The (111) case contains a global circulation that passes between the negative x, y, z tips and the positive x, y, z tips, circulations at each of the tips and a circulation in the root of the dendrite. The regions of high velocity occur in the global circulation and between the circulation in the root and the tips.

For well developed dendrites the thermoelectric currents can become very complex. Figure 5.52, shows the electric potential and direction of current density for a 3-dimensional dendrite with branches forming in the (111) direction as well as the (001) direction. Subsequently thermoelectric currents circulate from all the newly formed tips to local roots. It was only possible to calculate this result by removing the fluid dynamics solver, providing more computational resources to solve the electric potential, thus for this work the fluid dynamics that arise


Figure 5.42: Direction and magnitude of velocity field using the quasi 3D approximation



Figure 5.43: Direction of velocity field with magnetic field directed in the (001) direction



Figure 5.44: Magnitude of velocity field with magnetic field directed in the (001) direction



Figure 5.45: Direction of velocity field with magnetic field directed in the (011) direction



Figure 5.46: Magnitude of velocity field with magnetic field directed in the (011) direction



Figure 5.47: Direction of velocity field with magnetic field directed in the (111) direction



Figure 5.48: Magnitude of velocity field with magnetic field directed in the (111) direction



Figure 5.49: Direction of velocity field with magnetic field directed in the (123) direction



Figure 5.50: Magitude of velocity field with magnetic field directed in the (123) direction



Figure 5.51: Regions of high and low pressure for each orientation of the magnetic field. Top left: (001), Top right: (011), Bottom left: (111), Bottom right: (123)

from this situation will be not considered.

5.5 Effect of Magnetic Field on Crystal Morphology

The previous section demonstrated that a wide variety of flow fields is achievable by changing the orientation of the magnetic field. When considering morphological changes the magnitude of the flow field is also very important. This can be characterised by the magnitude of the local Péclet number:

$$Pe = Re \cdot Pr = \frac{uL_0}{\alpha} \tag{5.12}$$

The cases in the previous section assume that $Pe \ll 1$, such that the system is dominated by diffusion and convective transport has a negligible effect on the crystal morphology. In the case of supercooled dendritic growth the characteristic length L_0 is not easily quantified. For example in conventional casts it is standard practice to approximate the characteristic length as the diameter of the crystal ($O10^{-5}$ m) or the tip radius ($O10^{-6}$ m). However in the case of supercooled growth the thermoelectric currents are localised within the diffusion boundary layer ($O10^{-7}$ m) and if the interface destabilises leading to secondary branching then this will lead to thermoelectric currents circulating around the secondary arms in which case the secondary arm spacing ($O10^{-7}$ m) could be more suitable. Perturbations at the interface caused by micro fluid jets may even act on length scales smaller ($O10^{-8}$ m) This gives a range spanning over three orders of magnitude and it is likely that a suitable value of L_0 will also be dependent on both the material properties and the experimental set up. This large variety of L_0 coupled with a moving interface causes difficulties in deciding the



Figure 5.52: 3D stagnant growth of a well developed dendrite. Top: Potential, Bottom: Direction of current density

magnitude of Pe.

Assuming that the diffusion boundary layer is a good approximation then in conventional systems $L_0 \sim 010^{-5}$ m and in supercooled systems $L_0 \sim 010^{-7}$ m. There will also be a significant difference in the thermal gradients $\nabla T \sim 010^4 010^5 \text{ Km}^{-1}$ compared to $\nabla T \sim 010^7 - 010^8 \text{ Km}^{-1}$. However since Pe is dependent on L_0 and assuming that there is some scalability between length scales then in order to achieve a similar Peclet number in supercooled melts a significantly larger velocity is required. In order to achieve this a much larger magnetic field is required than may be necessary in practice, consequently the resistive forces may become significant and therefore must be taken into consideration.

Assuming the Reynolds number will exist in the region of creeping flow, then the viscous term in Navier-Stokes equation will be dominant. The total force can be approximated as a balance between the Lorentz force, the corresponding resistance force $(\mathbf{u} \times \mathbf{B} \times \mathbf{B})$, viscous forces and finally an effective resistance force which is introduced through the moving interface. In terms of dendritic growth consider a tip growing into the bulk, with a circulation of flow around it as predicted in the previous section. At some later time the dendrite tip will have moved through the circulation, which for this purpose can be considered to now be circulating around the trunk. The Lorentz force at the tip will then have to re-accelerate the flow in front of the tip and existing momentum will be directed against the Lorentz force. A qualitative representation of this is given in figure 5.53. In the moving reference frame of the tip this can be considered as an effective retarding force and to simplify the problem consider a perpendicular flow across a planar front which is solidifying at a velocity of u_{tip} and a perpendicular flow that exists purely in the boundary layer with a velocity \mathbf{u} as shown in figure



Figure 5.53: Qualitative representation of Lorentz force and momentum at the dendrite tip as it grows



5.54. To avoid including the dynamics of the circulation it is assumed that as the

Figure 5.54: 1D representation of estimating the effect of momentum loss through tip growth

solid front moves, there is no variation in density and that during solidification momentum is lost through structural bonds as the crystal lattice forms, this is analogous to the circulation passing the tip. The volumetric change in momentum due to this can be written as:

$$\frac{\mathbf{F}_{\text{eff}}}{V} = \rho \frac{\mathbf{u}}{t_0} \tag{5.13}$$

where the characteristic time t_0 is taken to be the time taken for the moving front to pass through some characteristic length (boundary layer):

$$t_0 = \frac{L_0}{u_{tip}} \tag{5.14}$$

this gives an effective volumetric force of:

$$\frac{\mathbf{F}_{\text{eff}}}{V} = \frac{\rho \mathbf{u} u_{tip}}{L_0} \tag{5.15}$$

Introducing the other forces into Newton's 2nd law of motion gives:

$$\rho \frac{d\mathbf{u}}{dt} = \sigma S \nabla T \times \mathbf{B} - \mu \nabla^2 \mathbf{u} - \sigma \mathbf{u} \times \mathbf{B} \times \mathbf{B} - \frac{\rho \mathbf{u} \cdot \mathbf{u}_{tip}}{L_0}$$
(5.16)

Assuming the Lorentz and viscous terms act purely in the direction of flow then:

$$\nabla^2 \mathbf{u} = \frac{d^2 u}{dy^2} \sim \frac{u}{L_0^2} \tag{5.17}$$

and Newton's 2nd law can be re-written as:

$$\rho \frac{du}{dt} = \sigma S \nabla T B - u \left(\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho u_{tip}}{L_0} \right)$$
(5.18)

Under steady state conditions the maximum velocity is given by:

$$u_{ss} = \frac{\sigma S \nabla TB}{\left(\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho u_{tip}}{L_0}\right)}$$
(5.19)

The magnetic field which gives the maximum velocity is given by:

$$\frac{du_{ss}}{dB} = 0 \tag{5.20}$$

giving:

$$B = \left(\frac{1}{\sigma} \left(\frac{\mu}{L_0^2} + \frac{\rho u_{ss} u_{tip}}{L_0}\right)\right)^{\frac{1}{2}}$$
(5.21)

Figure 5.55 shows the velocity normalised by the steady state velocity at $\frac{du_{ss}}{dB} = 0$ against the magnetic field strength for different length scales with a tip velocity of zero. For high magnetic field strengths the induced term becomes dominant and the flow is damped, but the magnitude of the magnetic field which gives



Figure 5.55: Normalised steady state velocity as a function of magnetic field for various length scales

a maximum velocity is highly dependent on the characteristic length; ranging from 0.5T - 50T. There is no dependence on $S\nabla T$ when determining the **B** field that will give the maximum velocity only the characteristic length scale and tip velocity. Figure 5.56 shows how the maximum velocity varies as a function of the corresponding length scale. When the effective resistance from the moving interface is included another shift in the maximum velocity can be deduced. Using a tip velocity of 20 ms⁻¹ as observed in supercooled dendritic growth, the shift is significant, moving the magnetic field somewhere between 8T - 100T.

This 1D model shows that the maximum convective effects should occur at some optimal value of **B**. However it is also necessary to include the transient term to see how long it takes to accelerate the fluid, as this may be an important factor if the flow is unable to reach its maximum at the optimal value for **B**.



Figure 5.56: Optimal magnetic field as a function of length scale.



Figure 5.57: Normalised steady state velocity as a function of magnetic field for various length scales with a tip velocity of $20\,\rm ms^{-2}$

Returning to Newton's 2nd Law and integrating by separation of variables:

$$\int dt = \rho \int \frac{du}{\sigma S \nabla T B - \left(\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}}\right) u}$$
(5.22)

$$t + c = -\frac{\rho}{\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}}} \ln\left(\sigma S\nabla TB - \left(\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}}\right)u\right)$$
(5.23)

using the condition u = 0 at t = 0 gives:

$$c = -\frac{\rho}{\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}}} \ln (\sigma S \nabla T B)$$

$$t = \frac{\rho}{\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}}} \left(\ln (\sigma S \nabla T B) - \ln \left(\sigma S \nabla T B - \left(\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}} \right) V \right) \right)$$
(5.24)
$$(5.25)$$

rearranging for u:

$$u = \frac{\sigma S \nabla T B}{\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}}} \left(1 - e^{-\frac{1}{\rho} \left(\frac{\mu}{L_0^2} + \sigma B^2 + \frac{\rho L_0}{u_{tip}}\right) t} \right)$$
(5.26)

Using a characteristic length $L_0 = 10^{-7}$ m and a variety of tip velocities figure 5.58 shows the time taken to accelerate to the maximum flow velocity. The velocities are normalised against the case where there is no moving tip, this is because only the magnitude of the velocity is dependent on $S\nabla T$ and not the time taken to accelerate to the maximum value. Thus the general form is applicable to any Seebeck number and thermal gradient at this length scale. Figure 5.58 highlights two important criteria for effectively modelling this phenomenon. The first is that the time to accelerate the flow is much larger than time step used in solidification and is comparable to the characteristic time. The second is that a moving tip has a significant impact on the maximum achievable velocity. Both of these effects show simplifying the problem to a set of steady state simulations is erroneous. For the first case a steady state approximation would significantly over



Figure 5.58: Normalised velocity accelerating with B = 10 T and $L_0 = 10^{-7}$ m

predict the early stages of growth and for the latter case it would be necessary to model the effective resistance force. Thus to model the full change in growth mechanics at this length scale it is chosen to run fully transient simulations. This model although somewhat simplistic also provides some operational parameters of magnetic field strengths that can be used. The maximum field used is 20T and although the true value of L_0 is not well known it is plausible that within this range the dampening terms will become negligible in regions of high Lorentz forces. However in regions of low Lorentz force, where fluid flow may still be large the resistive term is necessary and therefore is included in the model.

5.5.1 Fully Coupled

To explore the consequence of including convective transport the 2-dimensional model with quasi 3-dimensional boundary conditions is used. A magnetic field in the z-direction provides Lorentz forces in the plane of growth. Four cases are

presented here; two involving a pure material with a significant Seebeck power and two representing growth of a binary alloy with a Seebeck power representative of an Aluminium Silicon alloy. A 10T and a 20T magnetic field are applied to a pure material and the transient growth, the thermal field at the final time, the magnitude of \mathbf{J} , the direction of velocity and magnitude of velocity are given in figures 5.59, 5.60, 5.61, 5.62 and 5.63 respectively. The transient contours represent the interface at different times during the solidification process beginning at t = 2000 and then taking equal steps of $\Delta t = 4000$. In both cases the primary tip velocity is increased compared to stagnant conditions and the tip is rotated clockwise from the direction of preferential growth and this deflection is increased in the higher magnetic field case. It is important to note that this deflection is not physical rotation as Newtonian forces have been neglected and the crystal anisotropy is still directed along the computational axis. The interface can also be seen to destabilise and secondary branching occurs in the clockwise side of the primary arms and on the anti-clockwise side of the primary arms the thermal boundary layer is extended. All of these changes are a direct consequence of the convection introduced through the interaction of the thermoelectric currents and the magnetic field. The mechanism of these changes can be attributed to the dynamics seen in the low magnetic field strength approximation cases; in figure 5.42 using the quasi 3-dimensional approximations circulations at the tips and a global circulation around the dendrite were resolved. The circulation at the tip is responsible for the deflection; by providing an increase of free energy on the incident side of the tip. Over time this constant bias results in the tip deflecting to a constant angle. Hypothetically this can be treated as a modification to the tip equilibrium formed in microscopic solvability. Although a formal proof is beyond the scope of this thesis. Qualitatively, by introducing the convective effects, the equilibrium between the growth of the crystal tips and the amount of free energy will form at this deflected angle. The secondary branching can be attributed to both the global circulation and the tip circulation, where the incident flow onto the clockwise side of the arm perturbs the local free energy initiating secondary growth. As the global circulation passes over the crystal arm into the quasi 3-dimensional plane, hotter fluid is transported and deposited on the anticlockwise side of the arm extending the thermal boundary layer in this direction. The growth of secondary arms has a significant effect on the fluid dynamics; with secondary circulations forming at the tips of the secondary branches and based on a similar argument above for the primary tips a bias of free energy forms at the secondary tips and a deflection is observed. The global circulation is also shifted following the trunks of the secondary arms and although not directly observed in these results it is quite plausible that under certain circumstances the initiation of tertiary branches could form in a similar fashion to the secondary branches. The reason for the change to the fluid dynamics can be understood by looking at the current density given in figure 5.61, where the morphological changes to the dendrite have also caused a significant change to the location of high thermoelectric currents, which are now situated at the tips of the secondary branches as well as the primary branch. As secondary branches form the local curvature changes and through satisfying the equilibrium temperature at the interface the boundary condition for thermoelectric currents also changes. This can be seen in figure 5.64; the solved region is a locus around the interface and since the currents are localised close to the interface the solved region is large enough to capture the dynamics. The implications of these dynamics are clearly

shown when looking at the direction of the thermoelectric currents given in figure 5.65. Micro circulations of \mathbf{J} form around each of the secondary arms, essentially emanating from both the primary and secondary tips and terminating in a local root and not necessarily the primary root. The bottom part of figure 5.65 represents a close up image of the upper right quadrant of the dendrite and for clarity normalised vectors have been plotted. The circulations are quite notable and it is also possible to see current passing between arms creating complex circulation routes. A similar situation effectively occurs in all of the cases presented and so only the 20T case has been shown in detail. Figure 5.63 shows how these moving regions of high current in moderately high magnetic fields cause the regions of high fluid velocity to move with the tips of the dendrite. In both cases a circulation around the nucleation site exists, however the magnitude is higher in the 10T case. As the dendrite develops this region becomes devoid of thermoelectric currents as the temperature is almost isothermal and therefore the driving Lorentz force has also diminished. The only forces acting in this region are the viscous and more importantly the resistive forces. The resistive force is dependent on the magnitude of the magnetic field and therefore this circulation is more damped in the 20T case compared to the 10T case.

When investigating alloy solidification the only significant difference compared to a pure material is the interface equilibrium dependence on the local solute concentration. A difficulty arises when including realistic transport of solute, which for diffusion is dependent on the mass diffusivity. In real systems the Lewis number which is the ratio between the thermal diffusivity and mass diffusivity is typically around 1000, which introduces yet another significant disparity in time scales. In this work a Lewis number of 10 is used and although not exactly



Figure 5.59: 2D evolution of a pure material in the presence of a magnetic field. Top: $\mathbf{B} = 10$ T, Bottom: $\mathbf{B} = 20$ T



Figure 5.60: 2D temperature field of a pure material in the presence of a magnetic field. Top: $\mathbf{B} = 10$ T, Bottom: $\mathbf{B} = 20$ T



Figure 5.61: 2D magnitude of **J** for a pure material in the presence of a magnetic field. Top: $\mathbf{B} = 10$ T, Bottom: $\mathbf{B} = 20$ T



Figure 5.62: 2D direction of velocity for a pure material in the presence of a magnetic field. Top: $\mathbf{B} = 10$ T, Bottom: $\mathbf{B} = 20$ T



Figure 5.63: 2D magnitude of velocity for a pure material in the presence of a magnetic field. Top: $\mathbf{B} = 10$ T, Bottom: $\mathbf{B} = 20$ T



Figure 5.64: 2D electric potential for a pure material in the presence of a 20T magnetic field. Top: Regions of submesh, Bottom: Potential



Figure 5.65: 2D direction of \mathbf{J} for a pure material in the presence of a magnetic field. Top: whole dendrite, Bottom: upper right quadrant

representative of a real system it exhibits similar characteristics. A 3T and a 10T magnetic field are applied to a solidifying binary alloy that exhibits properties of an Aluminium Silicon alloy. The transient growth, the thermal field, the concentration field, the magnitude of \mathbf{J} , the direction of velocity and magnitude of velocity are given in figures 5.66, 5.67, 5.68, 5.69, 5.70 and 5.71 respectively. In both cases similar morphological changes occur compared to the case of a pure material and the mechanism is essentially identical. The most significant difference is the increased secondary branching and their initiation closer to the primary tip, which appears to be a consequence of the slower growth velocity of the tip, allowing a longer time for the interface to destabilise on the side of the primary arm. Using the same argument at the tip of the dendrite suggests that for a given magnetic field the deflection should increase in alloy solidification compared to a pure material. However although it is not obvious the deflection of the tip in the 10T alloy case is slightly less than the pure case, which could be a due to the solute deposition caused by the local secondary circulations near to the tip. This additional solute on the clockwise side of the tip will discourage the deflection and change the tip equilibrium making it dependent on the growth of secondary branches. This may prevent a steady state solution from existing and instead the equilibrium will oscillate as consecutive secondary branches form. Figure 5.71 shows velocity for both the alloy cases. In both cases the circulation around the nucleation site is still evident compared to the pure cases, however in the 3T case this circulation is larger compared to the velocity near the tips of the dendrite. Two fundamental differences are apparent from this case; the first is the dampening term is now significantly less compared to the other cases and thus the time taken to damp out the central circulation is much longer and

the second is the time taken to accelerate the flow at the tips is also significantly longer and the maximum velocity achievable is significantly less than the steady state solution. The contrast between the flow fields in the 3T and 10T alloy cases highlights the necessity to use time-dependent models.

By definition of the problem description the dendrite should exhibit rotational symmetry, however although not initially obvious the 20T pure material case in figure 5.59 exhibits a slight asymmetry, which can be seen by comparing the length of the secondary branches. Physically the problem is an unstable equilibrium and a single asymmetric perturbation can lead to unbounded growth removing any symmetry. In this case the perturbations occur as a consequence of numerical error in the velocity, introduced by the fluid dynamics solver. As the force increases the convergence criteria for continuity effectively decrease, compounded with the necessity of increasing relaxation parameters requires significantly more iterations to reach a fully converged solution increasing the computational time of the simulation. However the fluid dynamics solver does provide a very good estimate of the change in momentum for each time step and the error is likely to be significantly smaller than errors that occur through natural thermal fluctuations. It is unclear though if this error could introduce any artificial bias as any initial small difference affecting the liquid fraction and the thermoelectric currents, can only be noticed once the crystal develops. However this example shows that the calculations are performed across the full domain, this allows for multiple crystals to be simulated simultaneously when investigating grain growth. Preliminary results of this are given in section 7.2.

Up to this point the assumption that the quasi 3-dimensional boundary condition is applicable has been taken, although the flow field matches the 3-dimensional



Figure 5.66: 2D evolution of a binary alloy in the presence of a magnetic field. Top: $\mathbf{B} = 3T$, Bottom: $\mathbf{B} = 10T$



Figure 5.67: 2D temperature field of a binary alloy in the presence of a magnetic field. Top: $\mathbf{B} = 3T$, Bottom: $\mathbf{B} = 10T$



Figure 5.68: 2D solute concentration field of a binary alloy in the presence of a magnetic field. Top: $\mathbf{B} = 3T$, Bottom: $\mathbf{B} = 10T$



Figure 5.69: 2D magnitude of **J** for a binary alloy in the presence of a magnetic field. Top: $\mathbf{B} = 3T$, Bottom: $\mathbf{B} = 10T$


Figure 5.70: 2D direction of velocity for a binary alloy in the presence of a magnetic field. Top: $\mathbf{B} = 3T$, Bottom: $\mathbf{B} = 10T$



Figure 5.71: 2D magnitude of velocity for a binary alloy in the presence of a magnetic field. Top: $\mathbf{B} = 3T$, Bottom: $\mathbf{B} = 10T$

calculations, the corresponding growth may not necessarily, thus to test this a 3-dimensional simulation looks at the change in morphology. Currently it is extremely time consuming and memory intensive to simulate a 3-dimensional crystal that is large enough for any significant morphological changes to be observed and so the idea of a moving mesh is adopted and only the tip is considered. Figure 5.72 shows the development of the tip, initially no magnetic field is present and the crystal grows until microscopic solvability is satisfied. The mesh moves to keep the tip central to the domain in the x-direction, but does not correct for the y and z directions so any changes can be observed. The first figure is at time 4800 when the x tip reaches the centre of the domain and up to this point the mesh has not moved. The second figure is at time 6400 and the mesh has begun to move, this is visible as the y and z tips can be seen to be disappearing through the negative x face of the domain. The third figure is at time 20000, when the tip has reached microscopic solvability and both the thermal and liquid fraction fields are time independent, a magnetic field in the positive z-direction of 20T is now applied and the flow begins to accelerate. The fourth figure shows a bulge that has formed on the negative y side of the tip, this represents the initiation of a secondary branch, however as the tip continues to grow the information of how this secondary branch develops is lost. The branches continue to form and bumps appear to oscillate with each branch formed. The thermal field can also be seen to be extended in the positive y-direction. Figure 5.73 is at time 40000, where the negative y boundary conditions of the domain are starting to influence the solution. The tip can be seen to have deflected significantly away from the y = 0plane towards the negative y face. Similar morphological changes are observed in this situation as the 2-dimensional cases. It is plausible that the deflection could actually be greater, however the thermal and liquid fraction boundary condition on the negative x face of the domain influence the solution by providing information applicable to growth along the computational axis.



Figure 5.72: 3D Transient growth with a moving mesh. Top left: t = 4800, Top right: t = 6400, Bottom left: t = 20000, Bottom right: t = 33200

The mechanism of the growth is similar to the quasi 3-dimensional case, figure



Figure 5.73: 3D Transient growth with a moving mesh. Top: t = 39800, Bottom: Deflection from preferred direction of growth

5.74 shows the direction and magnitude of the velocity field. The circulation at the tip is clearly visible and the maximum velocity appears at the front of the tip. The other maximum velocity on the negative y side of the tip, marks the remnants of a secondary branch and its corresponding circulation. Flow incident on the tip introduces colder fluid lowering the free energy and causing the deflection and as it passes over the tip deposits warmer fluid on the far side of the tip extending the thermal layer.

5.6 Simulation Time

This section aims to provide a quantified estimation of the computational time required for solving the models presented in this thesis. The model comprises three solvers; solidification, electric potential and fluid dynamics. At the beginning of the solidification solver the extremes of the indices i, j, k are found based on the current liquid fraction, this provides a cube that contains all the cells required in the solidification algorithm and the rest of the domain can be ignored. However the majority of the solver's time is spent purely on the interfacial cells, which in 3dimensions can be considered to be an expanding sphere with a surface area that is increasing as Ot^2 and in 2-dimensions this becomes Ot. The electric potential in 3-dimensions is solved inside a shell that is defined via the interface and so this also has a computational expense of Ot^2 in 3-dimensions and Ot in 2-dimensions. Fluid dynamics are solved throughout the domain, however in the early stages of growth the far field velocities effectively satisfy conservation laws and so fewer iterations are required to resolve the local fluid mechanics. As the crystal grows, more iterations are required in 3-dimensions where the liquid is prevented from



Figure 5.74: Direction and magnitude of velocity in 3D growth with a moving mesh

entering the solid; the region of interest is essentially a shell around the dendrite and the number of iterations required increases as Ot^2 , however when using the quasi 3-dimensional approximation the fluid dynamics internal to the system are important and so the number of iterations for convergence may also increase as Ot^2 . Although largely dependent on the problem being solved typically 400 iterations are sufficient for a converged solution using values for momentum false time step = 0.1 and pressure under relaxation = 0.7. Solving a time step in 3-dimensions therefore takes significantly longer than in 2-dimensions, however the tip velocity in 3-dimensions is generally significantly faster and so fewer time steps are required. Table 5.1 shows typical simulation times for a single core running at approximately 2.5Ghz.

Simulation Type	2D	3D
Stagnant growth	2 hours	1-2 days
Stagnant growth with electric potential solver	12-24 hours	3-4 days
Forced convection	1-2 days	2-3 days
Growth with magnetic field	1-2weeks	n/a
Moving mesh growth with magnetic field	4 weeks	4-6 weeks
Grain growth	3-4 weeks	n/a

Table 5.1: Approximate time taken to preform various simulations

5.7 Summary

This chapter explored the implementation of the numerical model and the predictions that were made. Several cases were investigated each introducing an increased detail of the physical phenomena and for each case simulations are presented in both 2-dimensions and 3-dimensions. The first tests looked at a pure material with transport through diffusion to see how the dendritic model behaved. The second case introduced a directional forced convection to explore how modifications to the transport equations affect dendritic growth. The third case investigated the flow patterns that emerge when an external magnetic field is applied by changing the orientation of the magnetic field and using a low magnetic field strength approximation, which effectively causes the transport equations to be dominated by diffusion and so no convective morphological changes occur. The model was then fully coupled in 2-dimensions and corresponding changes to the crystal morphology from thermoelectric magnetohydrodynamics were investigated. A binary alloy was then introduced using material properties based on AlSi, as significant Seebeck powers do not exist for pure materials and again the morphological changes and solute distribution were calculated. Finally the moving mesh technique is used to qualify the quasi 3-dimensional approximation as similar morphological changes were seen in both.

Chapter 6

Theoretical Verification

6.1 Overview

In this chapter an analytic solution is constructed by simplifying the crystal morphology to a sphere. The electric potential then becomes the solution to Laplace's equation which is solved analytically through spherical harmonics. From this solution the current density and corresponding Lorentz forces can simply be calculated for any orientation of magnetic field. Introducing this force into a low magnetic field strength approximation numerical model reveals similar flow fields to those described in the Results chapter.

6.2 Analytic Solution for Lorentz Forces

Currently there are no experiments that address MHD flow at this length scale and with electrical currents of this form. The solidification experiments that do exist only show qualitative agreement to the observed morphological changes. Thus part of the validation of the work presented here addresses an analytic solution of a simplified problem. Based on the assumption that the crystal is spherical, there is a constant mean curvature everywhere on the interface and from the Gibb's Thompson condition in equation 2.21 the interfacial equilibrium temperature relative to the melting temperature is given by:

$$T' = T^i - T_m = -C\gamma(\theta, \phi) \tag{6.1}$$

where:

$$C = \frac{T_m \kappa}{L} \tag{6.2}$$

T' is not the real temperature, but purely scaled from the equilibrium temperature by constants. Taking γ to be the surface energy of the form $(N_x^4 + N_y^4 + N_z^4)$, the interfacial temperature mimics that of a dendrite. The Cartesian co-ordinates are transformed such that:

$$x = r \sin \theta \cos \phi$$

$$y = r \sin \theta \sin \phi$$
 (6.3)

$$z = r \cos \theta$$

where θ is the zenith angle and ϕ represents the azimuth angle. Thus in Spherical co-ordinates this becomes:

$$T' = -\cos^4\theta + \sin^4\theta \left(\cos^4\phi + \sin^4\phi\right) \tag{6.4}$$

A representation of this is given in figure 6.1, where the corresponding locations of the tips on an equiaxed dendrite are undercooled.

Performing the same steps as in section 2.2 through conservation of charge and assuming that S is non-varying with T gives the full solution to the potential as Laplace's equation:

$$\nabla^2(\Psi) = 0 \tag{6.5}$$



Figure 6.1: Analytic surface temperature on a sphere

Assuming that free surface charges will distribute in the same form as the surface energy and solving for $-\Psi_l$ the boundary condition at the interface becomes:

$$\Psi_{s}^{i} = \left(\frac{1}{2}\Delta S + \Psi_{E}^{i}\right)T$$

$$-\Psi_{l}^{i} = \left(\frac{1}{2}\Delta S - \Psi_{E}^{i}\right)T$$

(6.6)

The general solution to Laplace's is given by equation 6.7, a formal derivation is given in the Appendix.

$$\Psi = \sum_{l=0}^{\infty} \sum_{m=0}^{l} \left[\left(A_l^m r^l + \frac{B_l^m}{r^{l+1}} \right) P_l^m \left(\cos\theta \right) \left(C_m \cos m\phi + S_m \sin m\phi \right) \right]$$
(6.7)

From equation 6.6 the boundary condition at the surface of the sphere can be written as:

$$\Psi_s(r_0) = \left(\frac{1}{2}\Delta S + \Psi_E^i\right) \left(\cos^4\theta + \sin^4\theta \left(\cos^4\phi + \sin^4\phi\right)\right) \tag{6.8}$$

$$\Psi_l(r_0) = \left(\frac{1}{2}\Delta S - \Psi_E^i\right) \left(\cos^4\theta + \sin^4\theta \left(\cos^4\phi + \sin^4\phi\right)\right) \tag{6.9}$$

Expanding this in terms of $\sum_{m=0}^{\infty} \sum_{n=0}^{\infty} \cos m\theta \cos n\phi$:

$$\Psi_{s}(r_{0}) = \left(\frac{1}{2}\Delta S + \Psi_{E}^{i}\right) \left(\frac{21}{32} + \frac{1}{8}\cos 2\theta + \frac{7}{32}\cos 4\theta + \frac{3}{32}\cos 4\phi - \frac{1}{8}\cos 2\theta\cos 4\phi + \frac{1}{32}\cos 4\theta\cos 4\phi\right)$$
(6.10)
$$\Psi_{l}(r_{0}) = \left(-\frac{1}{2}\Delta S + \Psi_{E}^{i}\right) \left(\frac{21}{32} + \frac{1}{8}\cos 2\theta + \frac{7}{32}\cos 4\theta + \frac{3}{32}\cos 4\phi - \frac{1}{8}\cos 2\theta\cos 4\phi + \frac{1}{32}\cos 4\theta\cos 4\phi\right)$$
(6.11)

Comparing this to 6.7 it can be seen that all $S_m = 0$. By inspection the solution can be seen to be even in both l and m and with a maximum order of 4 and the general solution becomes:

$$\Psi = \sum_{l=0}^{\infty} \sum_{m=0}^{l} \left(A_l^m r^l + \frac{B_l^m}{r^{l+1}} \right) P_l^m \left(\cos \theta \right) C_m \cos m\phi \tag{6.12}$$

The relevant Legendre Polynomials with the corresponding expansions to match the Ψ boundary condition are:

$$P_0^0(\cos\theta) = 1$$

$$P_2^0(\cos\theta) = \frac{1}{2} (3\cos^2\theta - 1) = \frac{1}{4} + \frac{3}{4}\cos 2\theta$$

$$P_2^2(\cos\theta) = 3\sin^2\theta = \frac{3}{2} - \frac{3}{2}\cos 2\theta$$

$$P_4^0(\cos\theta) = \frac{1}{8} (35\cos^4\theta - 30\cos^2\theta + 3) = \frac{1}{64} (9 + 20\cos 2\theta + 35\cos 4\theta)$$

$$P_4^2(\cos\theta) = \frac{15}{2} (7\cos^2\theta - 1)\sin^2\theta = \frac{1}{16} (45 + 60\cos 2\theta - 105\cos 4\theta)$$

$$P_4^4(\cos\theta) = 105\sin^4\theta = \frac{1}{8} (315 - 420\cos 2\theta + 105\cos 4\theta)$$

As $r \to 0 \ \frac{B_l}{r^{l+1}}$ becomes singular and similarly as $r \to \infty \ A_l r^l$ becomes singular except for the term where l = 0. Due to the fixed boundary at the interface it is possible to split the problem into two: inside the sphere where:

$$\Psi = \sum_{l=0}^{\infty} \sum_{m=0}^{l} A_l^m r^l P_l^m \left(\cos\theta \right) C_m \cos m\phi$$
(6.13)

$$\Psi = \sum_{l=0}^{\infty} \sum_{m=0}^{l} \left(\frac{B_l^m}{r^{l+1}} \right) P_l^m \left(\cos\theta \right) C_m \cos m\phi \tag{6.14}$$

By absorbing the coefficient C_m into A_l^m and B_l^m , expanding the summations and substituting for the Legendre Polynomials it is possible to match terms of corresponding $\cos n\theta \cos m\phi$ with the boundary condition. This leads to the following set of equations inside the sphere:

.

$$\begin{pmatrix} 1 & \frac{r_0^2}{4} & 0 & \frac{9r_0^2}{64} & 0 & 0 \\ 0 & \frac{3r_0^2}{4} & 0 & \frac{5r_0^4}{16} & 0 & 0 \\ 0 & 0 & -\frac{3r_0^2}{2} & 0 & \frac{15r_0^4}{4} & 0 \\ 0 & 0 & 0 & \frac{35r_0^2}{64} & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{105r_0^4}{16} & 0 \\ 0 & 0 & 0 & 0 & 0 & \frac{105r_0^4}{8} \end{pmatrix} \begin{pmatrix} A_0^0 \\ A_2^2 \\ A_4^0 \\ A_4^2 \\ A_4^4 \end{pmatrix} = \left(\frac{1}{2}\Delta S + \Psi_E^i \right) \begin{pmatrix} \frac{21}{32} \\ \frac{1}{8} \\ 0 \\ \frac{7}{32} \\ 0 \\ \frac{1}{32} \end{pmatrix}$$
(6.15)

And outside:

$$\begin{pmatrix} 1 & \frac{1}{r_0} & \frac{1}{4r_0^3} & 0 & \frac{9}{64r_0^5} & 0 & 0\\ 0 & 0 & \frac{3}{4r_0^3} & 0 & \frac{5}{16r_0^5} & 0 & 0\\ 0 & 0 & 0 & \frac{-3}{2r_0^3} & 0 & \frac{15}{4r_0^5} & 0\\ 0 & 0 & 0 & 0 & \frac{35}{64r_0^5} & 0 & 0\\ 0 & 0 & 0 & 0 & \frac{105}{16r_0^5} & 0\\ 0 & 0 & 0 & 0 & 0 & \frac{105}{16r_0^5} & 0\\ 0 & 0 & 0 & 0 & 0 & 0 & \frac{105}{8r_0^5} \end{pmatrix} \begin{pmatrix} A_0^0\\ B_0^0\\ B_2^0\\ B_2^2\\ B_4^0\\ B_4^2\\ B_4^4 \end{pmatrix} = \left(\frac{1}{2}\Delta S - \Psi_E^i \right) \begin{pmatrix} \frac{21}{32}\\ \frac{1}{8}\\ 0\\ \frac{7}{32}\\ 0\\ \frac{1}{32}\\ 0\\ \frac{3}{32}\\ -\frac{1}{8} \end{pmatrix}$$
(6.16)

The solution for the co-efficients are:

$$\begin{pmatrix} A_0^0 \\ A_2^0 \\ A_2^2 \\ A_2^2 \\ A_4^0 \\ A_4^2 \\ A_4^2 \\ A_4^4 \\ A_4^4 \\ B_0^0 \\ B_0^0 \\ B_2^0 \\ B_2^0 \\ B_2^2 \\ B_2^2 \\ B_2^2 \\ B_4^0 \\ B_4^2 \\ B$$

Thus the solutions for inside and outside of the sphere are given respectively by:

$$\Psi_s = \left(\frac{1}{2}\Delta S + \Psi_E^i\right) \left[\frac{3}{5} + \frac{2r^4}{5r_0^4}P_4^0(\cos\theta) + \frac{r^4}{420r_0^4}P_4^4(\cos\theta)\cos4\phi\right]$$
(6.18)

$$-\Psi_l = \left(\frac{1}{2}\Delta S - \Psi_E^i\right) \left[\frac{3}{5} + \frac{2r_0^5}{5r^5}P_4^0\left(\cos\theta\right) + \frac{r_0^5}{420r^5}P_4^4\left(\cos\theta\right)\cos4\phi\right]$$
(6.19)

The current density is given by:

$$= \frac{\partial \Psi}{\partial r} \hat{\mathbf{r}}$$

$$\mathbf{J} = \nabla \Psi = \frac{1}{r} \frac{\partial \Psi}{\partial \theta} \hat{\theta}$$

$$= \frac{1}{r \sin \theta} \frac{\partial \Psi}{\partial \phi} \hat{\phi}$$
(6.20)

To quantify Ψ_E^i , the condition:

$$\mathbf{J}_{\mathbf{s}}\hat{n} = \mathbf{J}_{\mathbf{l}}\hat{n} \tag{6.21}$$

must be satisfied. The normal to a spherical interface is given purely by the radial component of the current density, which in terms of associated Legendre polynomials is given by:

$$\mathbf{J}_{\mathbf{s}}\hat{r} = -\left(\frac{1}{2}\Delta S + \Psi_{E}^{i}\right) 4 \left[\frac{2r^{3}}{5r_{0}^{4}}P_{4}^{0}\left(\cos\theta\right) + \frac{r^{3}}{420r_{0}^{4}}P_{4}^{4}\left(\cos\theta\right)\cos4\phi\right]$$
(6.22)

$$\mathbf{J}_{\mathbf{l}}\hat{r} = -\left(\frac{1}{2}\Delta S - \Psi_{E}^{i}\right) 5\left[\frac{2r_{0}^{5}}{5r^{6}}P_{4}^{0}\left(\cos\theta\right) + \frac{r_{0}^{5}}{420r^{6}}P_{4}^{4}\left(\cos\theta\right)\cos4\phi\right]$$
(6.23)

Equating $\mathbf{J}_{\mathbf{s}}\hat{\mathbf{r}}$ and $\mathbf{J}_{\mathbf{l}}\hat{\mathbf{r}}$ at r_0 gives $\Psi_E^i = \frac{\Delta S}{18}$ indicating that for this given morphological assumption surface charge is an inherent part of the system and can be quantified. Accounting for surface charge inside the sphere \mathbf{J} becomes:

$$-\frac{5\Delta S}{9} \left[\frac{r^3}{r_0^4} \left[\frac{1}{5} \left(35\cos^4\theta - 30\cos^2\theta + 3 \right) + \sin^4\theta\cos 4\phi \right] \right] \hat{r}$$
$$\mathbf{J}_{\mathbf{s}} = -\frac{5\Delta S}{9} \left[\frac{r^3}{r_0^4} \left[\cos\theta\sin\theta \left(-7\cos^2\theta + \cos 4\phi\sin^2\theta + 3 \right) \right] \right] \hat{\theta}$$
(6.24)
$$\frac{5\Delta S}{9} \left[\frac{r^3}{r_0^4} \left[\sin 4\phi\sin^3\theta \right] \right] \hat{\phi}$$

and outside:

$$-\frac{4\Delta S}{9} \left[\frac{r_0^5}{r^6} \left[\frac{1}{4} \left(35\cos^4\theta - 30\cos^2\theta + 3 \right) + \frac{5}{4}\sin^4\theta\cos4\phi \right] \right] \hat{\mathbf{r}}$$
$$\mathbf{J}_{\mathbf{l}} = \frac{4\Delta S}{9} \left[\frac{r_0^5}{r^6} \left[\cos\theta\sin\theta \left(-7\cos^2\theta + \cos4\phi\sin^2\theta + 3 \right) \right] \right] \hat{\theta}$$
(6.25)
$$-\frac{4\Delta S}{9} \left[\frac{r_0^5}{r^6} \left[\sin4\phi\sin^3\theta \right] \right] \hat{\phi}$$

Figure 6.2 shows the direction of \mathbf{J} , where the current emanates from the region of high potential and crossing the interface in the low potential region. This is analogous to the numerical results observed on a crystal, with the high potential regions representative of the crystal tips and the low potential regions representing the crystal root. The relative magnitude of \mathbf{J} is given in figure 6.3, where the maxima occur at the tips and roots. The direction and relative magnitudes are in good agreement with the results on a growing dendrite from figures 5.27 and 5.28.

Applying a magnetic field in the (001) direction; aligned to the crystallographic orientation along the z-direction in spherical polar co-ordinates can be represented as:

$$(B_z \cos \theta)\hat{r}$$

$$B_z = -(B_z \sin \theta)\hat{\theta}$$

$$(0)\hat{\phi}$$

$$(6.26)$$

The Lorentz forces are then given by:

$$(J_{\phi}B_{z}\sin\theta)\hat{r}$$

$$\mathbf{J}\times\mathbf{B} = (J_{\phi}B_{z}\cos\theta)\hat{\theta}$$

$$- (J_{r}B_{z}\sin\theta + J_{\theta}B_{z}\cos\theta)\hat{\phi}$$
(6.27)

where **J** is the same as in equations 6.24 and 6.25. Therefore it is possible to also calculate the Lorentz forces analytically and through either Euler rotations described previously in equations 5.10 and 5.11 or by describing B_x and B_y directly in spherical polars:

$$(B_x \sin \theta \cos \phi)\hat{r}$$

$$B_x = (B_x \cos \theta \cos \phi)\hat{\theta}$$

$$(B_x \cos \phi)\hat{\phi}$$

$$(B_y \sin \theta \sin \phi)\hat{r}$$

$$B_y = (B_y \cos \theta \sin \phi)\hat{\theta}$$

$$(-B_y \sin \phi)\hat{\phi}$$
(6.29)



Figure 6.2: Analytic direction of ${\bf J}$



Figure 6.3: Relative magnitude of ${\bf J}$

Thus the first primitive set of basis functions can be defined analytically.

With the magnetic field orientated in the (001) direction the Lorentz force will only act in the x - y plane; figure 6.4 shows the Lorentz forces acting with the magnetic field in the (001) direction. In the z = 0 plane a similar direction of Lorentz force is observed when compared to 2-dimensional crystal growth cases. Taking a plane further along the crystal the corresponding forces above and also below the crystal arms are resolved on the sphere. Finally approaching the top of the sphere the reversal in sense of the Lorentz forces is captured as well as the forces responsible for the inter arm circulations in the root. The relative magnitude of the force close to the interface of the sphere shows a good agreement with the calculations of a crystal. The maximum forces appear in front of the x - y tips and also at the poles in the z direction.

To investigate the corresponding flow that is generated by these forces a low magnetic field approximation is taken to avoid the non-linear effects in the flow, also the numerical model is slightly modified. Using the algorithm for the intersection of a cube and a sphere from section 4.6.1, a sphere with the same radius r_0 used in the analytic solution is created by using the filled volumetric proportion on the Cartesian grid. Then by introducing the analytic Lorentz force in each cell the steady state flow field is calculated. The velocity streamlines are given in figure 6.5 and features similar to those seen on a crystal are observed; a global circulation in the ϕ -direction, circulation around the position of the crystal tips, circulations in the root and a circulation around the z tips. The relative magnitude of the velocity is given in figure 6.6 and the maxima occur at and around the x - y tips, the global circulation and the circulation around the z tips agreeing on a qualitative level with the calculations performed in the crystal model.

A good agreement for the other orientations presented in the results chapter is also observed in this analytic solution. The results for the (011) orientation are given in figures 6.7, 6.8 and 6.9, for the (111) orientation in figures 6.10, 6.11 and 6.12 and for the (123) orientation in figures 6.13, 6.14 and 6.15. Finally isosurfaces representing regions of high and low pressure are given in figure 6.16. In all cases force and velocity fields produced through this analytic solution can be directly compared to the corresponding numerical results on dendritic growth. This solution highlights how the surface temperature anisotropy can be responsible for the generation of significant thermoelectric currents. By keeping all key parameters constant except for the surface energy a qualitative agreement with numerical results encompassing the crystal growth mechanics can be approximated by an analytic solution on a sphere. Given that the majority of this solution is analytic and only a small part involves numerics this also provides an initial validation that the calculations are accurate given the current physical approximations.

The existence of surface charge in this solution shows that this is may also be the case in real experiments. By assuming that $\Psi_E^i = 0$ in the numerical model the magnitude of the surface charge can be determined by the discontinuity in $\mathbf{J}\hat{n}$. However without knowledge of the exact position of the interface this becomes very difficult. The implications of surface charge and its magnitude may provide a crucial mechanism that is currently omitted from this work.

6.3 Summary

This chapter constructed an analytic solution by simplifying the crystal morphology to a sphere. Using the surface energy to be representative of the surface



Figure 6.4: Analytic direction of Lorentz force with the magnetic field in the (001) direction in different planes Top left: z = 0, Top right: $z = \frac{3}{5}$, Bottom left: $z = \frac{11}{10}$, Bottom right: Surface magnitude



Figure 6.5: Direction of velocity with the magnetic field in the (001) direction



Figure 6.6: Magnitude of velocity with the magnetic field in the (001) direction



Figure 6.7: Analytic direction of Lorentz force with the magnetic field in the (011) direction in different planes Top left: z = 0, Top right: $z = \frac{3}{5}$, Bottom left: $z = \frac{11}{10}$, Bottom right: Surface magnitude

6.3 Summary



Figure 6.8: Direction of velocity with the magnetic field in the (011) direction



Figure 6.9: Magnitude of velocity with the magnetic field in the (011) direction



Figure 6.10: Analytic direction of Lorentz force with the magnetic field in the (111) direction in different planes Top left: z = 0, Top right: $z = \frac{3}{5}$, Bottom left: $z = \frac{11}{10}$, Bottom right: Surface magnitude

6.3 Summary



Figure 6.11: Direction of velocity with the magnetic field in the (111) direction



Figure 6.12: Magnitude of velocity with the magnetic field in the (111) direction



Figure 6.13: Analytic direction of Lorentz force with the magnetic field in the (123) direction in different planes Top left: z = 0, Top right: $z = \frac{3}{5}$, Bottom left: $z = \frac{11}{10}$, Bottom right: Surface magnitude

6.3 Summary



Figure 6.14: Direction of velocity with the magnetic field in the (123) direction



Figure 6.15: Magnitude of velocity with the magnetic field in the (123) direction



Figure 6.16: Analytic regions of high and low pressure for different orientations of magnetic field Top left: (001), Top right: (011), Bottom left: (111), Bottom right: (123)

temperature the electric potential can be solved and from this solution the current density and corresponding Lorentz forces can simply be calculated for any orientation of magnetic field. Introducing this force into a low magnetic field strength approximation numerical model reveals similar flow fields described in the results chapter. The analytic solution also indirectly showed the existence of surface charge at the interface; the implications of this however are not explored in this work.

Chapter 7 Conclusions and Future Work

7.1 Conclusions

The objective of this research was to address the following questions:

What are the effects on microstructural evolution due to an externally applied magnetic field during dendritic solidification?

What is the mechanism that causes change to dendritic morphology?

To answer these questions it was first necessary describe a dendritic model in 3-dimensions. This was accomplished by demonstrating that the enthalpy based method, which so far has only been used in 2-dimensional models was extendible into 3-dimensions and the implementation of this followed analytic trends consistent with the theory. Using a simple case involving forced convection, where there are already significant contributions from other authors, the morphological changes that occur through convective transport in the enthalpy method were
shown to be qualitatively comparable with the current literature. With these test cases providing reasonable results it was then possible to investigate the influence of magnetohydrodynamics.

This research has provided an insight into the complex dynamics that occur when an external magnetic field is applied to a solidifying alloy. By coupling together three fundamentally different physical fields (solidification, thermoelectricity and magnetohydrodynamics) into a numerical model the results presented show a plausible explanation for the mechanism that is occurring and the consequence this mechanism has on the overall dendritic formation. The theoretical results shown in Chapter 4 indicate that the Lorentz forces generated through the interaction of thermoelectric currents and an external magnetic field generate a complex flow structure comprising of many circulations. The nature of the flow is also shown to be dependent on the relative orientation of the magnetic field and the direction of dendritic growth. In 3-dimensions this provides a vast number of possible flow fields. Using a low magnetic field strength approximation, where it is assumed that the Lorentz forces perfectly balance the viscous force, the possible flow fields on a symmetric dendrite are characterised in the (001), (011) and (111) directions. This approximation causes both the Lorentz forces and velocities to become linear and the principle of linear superposition can be applied allowing for the fluid dynamics in any orientation to be calculated from a set of principal basis functions; the simplest being those of (001), (010) and (100). A further simplification also occurs by exploiting the symmetry of the dendrite morphology, both rotational and reflection, allowing for only one principal magnetic field direction to be simulated. This was chosen to be the (001)direction, then by applying Euler rotations the corresponding (010) and (100) directions can be simply calculated forming the principal basis functions. In all cases circulations form at the dendrite tips and a global circulation forms that envelops the dendrite entirely.

Under certain conditions; when the thermal gradients are large, the Seebeck power is large and the magnetic field is in a certain range, the velocity can become large enough to cause convective transport of solute and heat to become comparable to diffusion. This causes a redistribution of solute and heat causing a change in the local free energy to the liquid solid interface of the dendrite. In supercooled conditions the boundary layer for both the solute and thermal fields is very thin and in general the incident flow will provide favourable conditions to encourage growth. As flow passes over the dendrite the fluid becomes solute rich and hot on the opposite side to the incident flow. This causes a hot and enriched region where the local free energy decreases and the dendrite's growth is stunted. Using the quasi 3-dimensional model two significant morphological changes occur; the first is a deflection of the dendrite tip and the second is the initiation of secondary branching into the incident flow. The first morphology change is caused by circulations at the tips of the dendrite; the circulations continuously provide a region of higher free energy on the incident side while lowering it on the other, the net effect is a bias of growth in the direction of incident flow. This deflection is purely through the growth mechanics as Newtonian forces are neglected in this study and therefore the relative crystallographic orientation remains unchanged throughout the simulation. The second morphology change follows a similar argument, except that as they form down stream of the primary tip it is instead the action of the global circulation that influences this. The global circulation destabilises the interface on the incident side of the dendrite arm encouraging the growth of secondary branching. As the dendrite morphology changes due to these effects, the secondary branches form micro circulations of current, which in turn alter the Lorentz forces that are acting causing a change to the velocity field and circulations form at the secondary tips.

7.2 Future Work

This work has provided initial predictions of the expected morphological changes that could be expected when a magnetic field is applied to a solidifying alloy. The work thus far is purely theoretical and is based only on implementing reasonable approximations for numerous physical aspects that control solidification under these conditions. So far no experiment has been designed explicitly to observe the effects described in this work. Although this thesis provides an explanation for the observed phenomena, it is still somewhat qualitative and is far from being able to predict changes to overall material properties that may occur during this process. Given that morphological changes have been observed in practice and a possibility exists that these can be controlled by appropriate manipulation of the magnetic field, it would be beneficial to the material science community to design experiments to attempt to observe these phenomena directly.

Although the numerical errors associated with the solution for the velocity field from the Lorentz force derived from the analytic solution on a sphere should be insignificant, solving the Stokes equation rigorously would provide a way of reasonably calculating estimates of the fluid dynamics for any set of conditions. The analytic solution also indicates that surface charge may be an inherent consequence of the thermoelectric effect which will also interact with the electromagnetic field. Given that the surface charge is acting purely on the liquid solid interface, this could generate forces directly on the interface altering the surface tension and changing the growth mechanics.

Lorentz forces are generated in both the solid and liquid, for this study the effect of the solid forces has been neglected as it has no influence on the fluid dynamics. However for very high magnetic field strength in conventional casting systems, it is possible the situation will arise where the fluid dynamics is severely damped by the resistive term. In the solid region this could lead to significant stresses forming and if these are larger than the ultimate tensile strength of the material, the dendrite tips may shear off.

7.2.1 Grain Growth in a Magnetic Field

Preliminary work by Kao *et. al* [88] has been done on the influence of thermoelectric magnetohydrodynamics on grain structures. The parameters used were non-representative of any real material, but were intended to simply explore the various aspects of grain growth in a magnetic field. Two results are presented, both of a pure material, one with a high magnetic field and one with a moderate magnetic field. In contrast to the single crystal results (presented in the results section and assuming a linear scalability for all properties with respect to the deflection angle), the properties used in these results would correspond to somewhere in the range of 1T and 20T for the moderate and high magnetic fields respectively. The results for the transient evolution, final thermal field and velocities are given in figures 7.1, 7.2, 7.3 and 7.4. A deflection of the primary tip is observed in the high magnetic field case and in both cases secondary branching is increased in a similar fashion to the single growth presented earlier. An interesting feature of these simulations is the competition between dendrites to release latent heat; the central one is stunted due to this. These preliminary results also show that each dendrite initially acts independently and as they grow both their velocity fields and thermoelectric currents interact with other neighbouring dendrites. The flow field that emerges consists of a global circulation around all of the dendrites, with each tip retaining it's own circulations.

An interesting progression of this study would be to investigate the effect on multiple crystals in 3-dimensions, where as previously shown the orientation of the magnetic field plays a major role in the flow field that develops, in a similar fashion the interaction of randomly orientated crystals with respect to one another and the magnetic field may highlight other aspects of the overall dynamics taking place; some of which may be lost through the approximations taken when reducing the problem from 3-dimensions to 2-dimensions. This would involve significantly larger computational power and memory than has been used in producing the results for this study, but is well within the feasible limit of a moderate CPU cluster.

7.2.2 Peltier and Thompson Effects

For liquid metals the Seebeck effect is generally dominant over the Peltier and Thompson effects [1] and for this work the Peltier and Thompson effects have been neglected; simplifying the numerical model allowing for a more detailed analysis of the the transport through magnetohydrodynamics. However in the context of supercooled liquid metal solidification it is plausible that under cer-



Figure 7.1: Evolution of a grain in the presence of a magnetic field Top: Moderate magnetic field, Bottom: High magnetic field



Figure 7.2: Thermal field of grain growth in the presence of a magnetic field Top: Moderate magnetic field, Bottom: High magnetic field



Figure 7.3: Velocity of grain growth in a moderate magnetic field. Top: Direction, Bottom: Magnitude





Figure 7.4: Velocity of grain growth in a high magnetic field. Top: Direction, Bottom: Magnitude

tain conditions these effects may contribute significantly towards controlling the solidification mechanics. The details of these two effects are described in the theory section; in summary both effects involve the thermodynamically reversible transport of thermal energy via current carrying moving charges. The Thompson effect describes the volumetric heat transfer, while the Peltier effect is responsible for the heat transfer due to splitting and recombination of electron-hole pairs at the interface. The Thompson effect is controlled by variations in the Seebeck coefficient with position and is given by:

$$W = \frac{\mathbf{J}^2}{\sigma} - \mathbf{J}T \cdot \nabla S \tag{7.1}$$

while the Peltier effect is determined by the discontinuity in the Seebeck coefficient at the interface and is related to the Seebeck power by:

$$Q = r_c \mathbf{J}^2 \hat{n} + \Delta ST \mathbf{J} \hat{n} \tag{7.2}$$

In both equations the first term on the right is the thermodynamically irreversible joule heating. To compare the relative magnitude of each effect consider a 1-dimensional model of a moving tip with a constant velocity and a thermoelectric current emanating with a magnitude of $O10^{10}$ Am⁻². Under stagnant flow conditions the thermal transport is purely through diffusion. The generation of free energy to allow for the solidification front to release latent heat is governed by the thermal flux at the interface which can be written as:

$$Q = K \frac{dT_s}{dn} - K \frac{dT_l}{dn}$$
(7.3)

where T_s represents the temperature on the solid side and T_l represents the temperature on the liquid side. Assuming the thermal gradient on the solid side of

the interface is neglible then the flux is a function of the normal thermal gradient on the liquid side of the interface. The release of latent heat as the solid front moves can therefore be described as:

$$Lu_{tip} = K \frac{dT_l}{dn} \tag{7.4}$$

Using a tip velocity of $u_{tip} \sim 010 \,\mathrm{ms}^{-1}$ as predicted in the stagnant growth section and a typical volumetric latent heat $L \sim 010^9 \,\mathrm{Jm}^{-3}$ the thermal flux through diffusion at the interface $K \frac{dT_l}{dn} \sim 010^{10} \,\mathrm{Jm}^{-2} \mathrm{s}^{-1}$. This value can then be used to estimate the relative magnitude of joule heating, the Peltier effect and the Thompson effect to diffusion driven cases. To compare the volumetric joule heating term it is necessary to define the length scale over which it acts. Assuming this acts over the length of the thermal boundary layer ($L_0 \sim 010 \,\mathrm{m}^{-7}$) then using typical values the ratio of the diffusion to joule heating can be calculated by:

$$\frac{\frac{L_0 \mathbf{J}^2}{\sigma}}{K \frac{dT_l}{dn}} \sim 10^{-4} \tag{7.5}$$

which is sufficiently small to be neglected. A similar argument follows for the joule heating at the interface, with the exception that an approximation for the contact resistance needs to be taken. Assuming the resistivity varies linearly with distance and given that the interface thickness can be taken to be $L_0^i = O10^{-9}$ m then the contact resistance in terms of the conductivity becomes:

$$r_c = \frac{1}{\sigma} L_0^i \sim 10^{-17} \Omega$$
 (7.6)

giving a relative magnitude to diffusion of $O10^{-7}$. In this work it has been assumed that the Seebeck coefficient is non-varying inside the solid or liquid and only a discontinuity exists at the interface. From this definition $\nabla \cdot S = 0$ and the Thompson effect is non-existent, where solute is ejected during solidification a gradient of composition will form and a gradient of the Seebeck coefficient will exist. Using AlSi as an example with $T \sim 10^3$ K and assuming that a thin layer of pure silicon exists on the solid side of the interface, which then diffuses into the bulk and that the gradient of the Seebeck coefficient acts over the same length as the Thompson effect then the relative magnitude to diffusion can be approximated by:

$$\frac{\mathbf{J}T \cdot \nabla SL_0}{K\frac{dT_l}{dn}} \sim 0.4 \tag{7.7}$$

which on the onset seems comparable to diffusion. However given the localisation of the current density to the interface this value will be significantly lower even a small distance from the interface. At the interface the same procedure can be applied to the Peltier effect giving a ratio of:

$$\frac{\Delta ST \mathbf{J}\hat{n}}{K \frac{dT_l}{dn}} \sim 0.4 \tag{7.8}$$

These results show that the ratio of the Peltier effect with diffusion and the Thompson effect with diffusion are similar. This is perhaps expected with the approximations taken, essentially demonstrating that in regions approaching the interface the two effects become analogous. In reality moving away from the interface the gradient of the Seebeck coefficient is likely to decrease further restricting the Thompson effect. For this reason and the reduction in current density away from the interface it is possible to conclude that in general for thermoelectric currents in free dendritic growth the Peltier effect will be dominant over the Thompson effect, except in regions close to the interface, which for all intents and purposes can be approximated via the Peltier effect. Thus for the purpose of this hypothetical analysis the corresponding predictions will be constructed only in terms of the Peltier effect.

The ratio of 0.4 indicates that the Peltier effect may have a significant impact on the solidification mechanics; the root of the dendrite will become cooler and the tip will become hotter in comparison to a non-conducting dendrite with otherwise the same material properties. This change in temperature will cause the tip velocity to decrease and provide additional free energy for solidification in the root; the net effect is a coarsening of the dendrite. Consequently the change in interface temperature as the curvature changes will result in a reduction in the magnitude of the current density, which in turn will reduce the effect of the Peltier term. However predicting the change to the equilibrium state of the system is not possible due to the fully coupled nature. In the presence of a magnetic field, similar flow fields as presented in the result section will begin to form. The reduction in the current density will cause a decrease in the impulse in the liquid, however with a reduction in the tip velocity from the Peltier effect the flow will have longer to accelerate and interact. The results so far have shown that the fluid flow generally is at a maximum in regions of high current, which will also change the thermal transport in this region. Thus a more realistic solution would be achieved by coupling diffusion, convection and the Peltier effect and would provide an interesting continuation of the model presented in this thesis.

Appendix A

Derivations

This appendix provides a detailed derivation for some of the less obvious solutions

A.1 Calculation of Curvature

The gradient of a scalar f in Cartesian co-ordinates is given by:

$$\nabla f = \frac{\partial f}{\partial x}, \frac{\partial f}{\partial y}, \frac{\partial f}{\partial z}$$
(A.1)

-

with a magnitude of:

$$|\nabla f| = \left[\left(\frac{\partial f}{\partial x} \right)^2 + \left(\frac{\partial f}{\partial y} \right)^2 + \left(\frac{\partial f}{\partial z} \right)^2 \right]^{\frac{1}{2}}$$
(A.2)

Thus the normal unit vector of f is given by:

$$\hat{f} = \frac{\nabla f}{|\nabla f|} = \frac{\frac{\partial f}{\partial x}, \frac{\partial f}{\partial y}, \frac{\partial f}{\partial z}}{\left[\left(\frac{\partial f}{\partial x}\right)^2 + \left(\frac{\partial f}{\partial y}\right)^2 + \left(\frac{\partial f}{\partial z}\right)^2\right]^{\frac{1}{2}}}$$
(A.3)

The mean curvature can defined as:

$$2\kappa = \nabla \cdot \hat{f} = \nabla \cdot \frac{\nabla f}{|\nabla f|} \tag{A.4}$$

To avoid over complicating the equations let:

$$f_x = \frac{\partial f}{\partial x}, f_y = \frac{\partial f}{\partial y}, f_z = \frac{\partial f}{\partial z}$$
 (A.5)

for the first derivatives:

$$f_{xx} = \frac{\partial^2 f}{\partial x^2}, f_{yy} = \frac{\partial^2 f}{\partial y^2}, f_{zz} = \frac{\partial^2 f}{\partial z^2}$$
(A.6)

for the second derivatives and the cross terms:

$$f_{xy} = \frac{\partial^2 f}{\partial x \partial y}, f_{yz} = \frac{\partial^2 f}{\partial y \partial z}, f_{xz} = \frac{\partial^2 f}{\partial x \partial z}$$
(A.7)

The mean curvature can then be re-written as:

$$2\kappa = \frac{\partial}{\partial x} \left[\frac{f_x}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}} \right] + \frac{\partial}{\partial y} \left[\frac{f_y}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}} \right] + \frac{\partial}{\partial z} \left[\frac{f_z}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}} \right]$$
(A.8)

Applying the quotient rule of derivatives gives:

$$\frac{f_{xx} \left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}} - f_x \left[\frac{f_x f_{xx} + f_y f_x y + f_z f_{xz}}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}}\right]}{f_x^2 + f_y^2 + f_z^2}$$

$$2\kappa = + \frac{f_{yy} \left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}} - f_y \left[\frac{f_y f_{yy} + f_x f_x y + f_z f_{yz}}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}}\right]}{f_x^2 + f_y^2 + f_z^2}$$

$$+ \frac{f_{zz} \left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}} - f_z \left[\frac{f_z f_{zz} + f_x f_x z + f_y f_{yz}}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}}\right]}{f_x^2 + f_y^2 + f_z^2}$$
(A.9)

which is equivalent to:

$$2\kappa = \frac{1}{f_x^2 + f_y^2 + f_z^2} \left[\frac{\left(f_x^2 + f_y^2 + f_z^2\right) \left(f_{xx} + f_{yy} + f_{zz}\right)}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}} - \frac{\left(f_x^2 f_{xx} + f_y^2 f_{yy} + f_z^2 f_{zz} + 2f_x f_y f_{xy} + 2f_x f_z f_{xz} + 2f_y f_z f_{yz}\right)}{\left(f_x^2 + f_y^2 + f_z^2\right)^{\frac{1}{2}}} \right]$$
(A.10)

rearranging and cancelling terms gives the final form in 3-dimensions:

$$\kappa_{3D} = \frac{1}{2} \frac{(f_{yy} + f_{zz})f_x^2 + (f_{xx} + f_{zz})f_y^2 + (f_{xx} + f_{yy})f_z^2 - 2f_x f_y f_{xy} - 2f_x f_z f_{xz} - 2f_y f_z f_{yz}}{(f_x^2 + f_y^2 + f_z^2)^{\frac{3}{2}}}$$
(A.11)

In 2-dimensions the derivatives in the z-direction are zero including the cross terms:

$$f_z = f_{zz} = f_{yz} = f_{yz} = 0 (A.12)$$

by substitution gives:

$$\kappa_{2D} = \frac{1}{2} \frac{f_{yy} f_x^2 + f_{xx} f_y^2 - 2f_x f_y f_{xy}}{(f_x^2 + f_y^2)^{\frac{3}{2}}}$$
(A.13)

A.2 General Solution to Laplace's Equation in Spherical Polar Co-ordinates

In 3D spherical polar co-ordinates Laplace's can be written as:

$$\frac{\partial^2 \Psi}{\partial r^2} + \frac{2}{r} \frac{\partial \Psi}{\partial r} + \frac{\cos\theta}{r^2 \sin\theta} \frac{\partial \Psi}{\partial \theta} + \frac{1}{r^2} \frac{\partial^2 \Psi}{\partial \theta^2} + \frac{1}{r^2 \sin^2\theta} \frac{\partial^2 \Psi}{\partial \phi^2} = 0$$
(A.14)

Assuming that separation of variables can be applied such that:

$$\Psi(r,\theta,\phi) = R(r)\Theta(\theta)\Phi(\phi) \tag{A.15}$$

Substitution of Ψ into A.14 gives:

$$r^{2}\Theta\Phi\frac{\partial^{2}R}{\partial r^{2}} + 2r\Theta\Phi\frac{\partial R}{\partial r} + \frac{\cos\theta}{\sin\theta}R\Phi\frac{\partial\Theta}{\partial\theta} + R\Phi\frac{\partial^{2}\Theta}{\partial\theta^{2}} + \frac{1}{r^{2}\sin^{2}\theta}R\Theta\frac{\partial^{2}\Phi}{\partial\phi^{2}} = 0 \quad (A.16)$$

Assuming that Ψ is non-zero then dividing through by Ψ gives:

$$\frac{r^2}{R}\frac{\partial^2 R}{\partial r^2} + \frac{2r}{R}\frac{\partial R}{\partial r} + \frac{\cos\theta}{\Theta\sin\theta}\frac{\partial\Theta}{\partial\theta} + \frac{1}{\Theta}\frac{\partial^2\Theta}{\partial\theta^2} + \frac{1}{\Phi\sin^2\theta}\Theta\frac{\partial^2\Phi}{\partial\phi^2} = 0$$
(A.17)

Multiplying through by $\sin^2\theta$ and rearranging by collecting terms of $r,\,\theta$ and ϕ :

$$\left(\frac{r^2 \sin^2 \theta}{R} \frac{\partial^2 R}{\partial r^2} + \frac{2r \sin^2 \theta}{R} \frac{\partial R}{\partial r}\right) + \left(\frac{\sin \theta \cos \theta}{\Theta} \frac{\partial \Theta}{\partial \theta} + \frac{\sin^2 \theta}{\Theta} \frac{\partial^2 \Theta}{\partial \theta^2}\right) + \frac{1}{\Phi} \frac{\partial^2 \Phi}{\partial \phi^2} = 0$$
(A.18)

The solution to final term of A.18 is simple oscillator, with general solution:

$$\frac{1}{\Phi}\frac{\partial^2 \Phi}{\partial \phi^2} = -m^2 \tag{A.19}$$

Where the right hand side m is a constant. Substituting this back into A.18:

$$\left(\frac{r^2 \sin^2 \theta}{R} \frac{\partial^2 R}{\partial r^2} + \frac{2r \sin^2 \theta}{R} \frac{\partial R}{\partial r}\right) + \left(\frac{\sin \theta \cos \theta}{\Theta} \frac{\partial \Theta}{\partial \theta} + \frac{\sin^2 \theta}{\Theta} \frac{\partial^2 \Theta}{\partial \theta^2} \frac{1}{\Phi} \frac{\partial^2 \Phi}{\partial \phi^2}\right) = -m^2$$
(A.20)

Applying separation f variables again between r and θ :

$$\left(\frac{r^2}{R}\frac{\partial^2 R}{\partial r^2} + \frac{2r}{R}\frac{\partial R}{\partial r}\right) = l\left(l+1\right)$$
(A.21)

$$r^{2}\frac{\partial^{2}R}{\partial r^{2}} + 2r\frac{\partial R}{\partial r} - l\left(l+1\right)R = 0$$
(A.22)

Equation A.22 is Euler's differential equation with the general solution:

$$R = \sum_{n=0}^{\infty} a_n r^{n+c} \tag{A.23}$$

Substituting into A.22:

$$\sum_{n=0}^{\infty} (n+c) (n+c-1) a_n r^{n+c} + \sum_{n=0}^{\infty} 2 (n+c) a_n r^{n+c} - \sum_{n=0}^{\infty} l (l+1) a_n r^{n+c} = 0$$
(A.24)

$$\sum_{n=0}^{\infty} \left[(n+c)\left(n+c-1\right) + 2\left(n+c\right) - l\left(l+1\right)a_n r^{n+c} \right] = 0$$
 (A.25)

This must be true for all powers of r, where the most primitive form will be when n = 0, substituting this back into A.25 gives:

$$c(c+1) = l(l+1)$$
 (A.26)

$$c = l, -(l+1)$$
(A.27)

Thus the general form for R is:

$$R = \sum_{l=0}^{\infty} \left(A_l r^l + \frac{B_l}{r^{l+1}} \right) \tag{A.28}$$

Substituting the constant part back into A.22 gives:

$$l(l+1) + \frac{1}{\sin^2\theta} \left(\frac{\sin\theta\cos\theta}{\Theta} \frac{\partial\Theta}{\partial\theta} + \frac{\sin^2\theta}{\Theta} \frac{\partial^2\Theta}{\partial\theta^2} \right) = 0$$
 (A.29)

$$\frac{\partial^2 \Theta}{\partial \theta^2} + \frac{\cos \theta}{\sin \theta} \frac{\partial \Theta}{\partial \theta} + \left(l \left(l+1 \right) - \frac{m^2}{\sin^2 \theta} \right) \Theta = 0 \tag{A.30}$$

By inspection we can see that A.30 is of the form of an associated Legendre differential equation, with the solution of associated Legendre polynomials $P_l^m(\cos\theta)$. Thus combining all the terms together gives a general solution for Ψ as:

$$\Psi = \sum_{l=0}^{\infty} \sum_{m=0}^{l} \left[\left(A_l^m r^l + \frac{B_l^m}{r^{l+1}} \right) P_l^m \left(\cos \theta \right) \left(C_m \cos m\phi + S_m \sin m\phi \right) \right]$$
(A.31)

Appendix B

Publications Produced by this Research

- A. Kao, K. Pericleous, M. K. Patel and V. Voller, Effects of magnetic fields on crystal growth. International Journal of Cast Metals Research, 22 (1-4). pp. 147-150. ISSN 1364-0461 (2009)
- A. Kao, G. Djambazov, K. Pericleous and V. Voller, Thermoelectric MHD in dendritic solidification. Magnetohydrodynamics, 45 (3). pp. 305-315. ISSN 0024-998X (2009)
- 3. A. Kao, K. Pericleous, M. K. Patel and V. Voller, Thermoelectric MHD effects on equiaxed crystal morphology. In: 6th International Conference on Electromagnetic Processing of Materials, EPM 2009. Forschungszentrum Dresden-Rossendorf, Dresden, Germany. ISBN Forschungszentrum Dresden-Rossendorf (2009)
- 4. A. Kao, K. Pericleous, M. K. Patel and V. Voller, Thermoelectric effects

on alloy solidification microstructure. In: Modeling of Casting, Welding, and Advanced Solidification Processes - XII (MCWASP XII). The Minerals, Metals & Materials Society, Warrendale, Pennsylvania, USA, pp. 521-528. ISBN 978-0-87339-742-1 (2009)

- A. Kao, G. Djambazov, K. Pericleous and V. Voller, Effects of magnetic fields on crystal growth. In: Proceedings of the 7th International PAMIR Conference: Fundamental and Applied MHD and COST P17 Annual Workshop 2008. Institute of Physics, University of Latvia, Salaspils, Latvia, pp. 621-625. (2008)
- 6. A. Kao, K. Pericleous, M. K. Patel and V. Voller, The effects of thermoelectrically induced convection in alloy solidification. In: Proceedings of the Sixth International Conference on Engineering Computational Technology. Civil-Comp Press, Stirling, Scotland. ISBN 978-1-905088-26-3 (2008)
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