A NUMERICAL INVESTIGATION OF DIRECTIONAL BINARY ALLOY SOLIDIFICATION PROCESSES USING A VOLUME-AVERAGING TECHNIQUE

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A numerical investigation of directional binary alloy solidification processes is presented. In particular, a mathematical model is developed to study macrosegregation patterns as a consequence of thermal-solutal convection in the melt and mushy zone. A good understanding of the basic mechanisms of macrosegregation is helpful in designing and controlling solidification processes in order to achieve better quality cast products.

A volume-averaging technique is used to derive the macroscopic conservation equations for momentum, energy and species transport. This set of governing equations is applicable to the solid, mushy and liquid regions of the solidification system. Hence, the analysis is performed in a single-domain with a fixed numerical grid and a single set of boundary conditions.

Three finite element schemes for the solution of the momentum equation have been implemented. These schemes include a stabilized Galerkin equal-order-interpolation algorithm, a penalty function formulation and the fractional step method. Whereas the penalty method appears to yield the best efficiency, the fractional step method was shown to be the most reliable and hence is used for all the simulations in the present study.

The numerical model is developed in two stages. First, the set of macroscopic conservation equations is applied to natural convection and double diffusive con-
vection problems in porous media. The simulation of fluid flow through porous media could help validate the application of the volume-averaging equations to solidification process modelling where the mushy region is typically considered as a porous medium. The numerical results are compared with various porous media flow predictions reported in the literature. Good agreement with the results of earlier studies shows that the macroscopic equations derived through the volume-averaging technique are indeed valid in both single- and two-phase regions.

Secondly, the above model is applied to the simulation of directional binary-alloy solidification processes. The developed single-domain model is shown to predict well the extended mushy zone and the formation of channels. It is found that the variation of the alloy composition in the cast product reaches 40% of the initial composition. This finding supports the theory that thermal-solutal convection can induce severe macrosegregation. The effects of anisotropic permeability of the mushy zone on the obtained segregation patterns are also studied. A simulation of pure aluminum solidification is also carried out as a limiting case to test the versatility of the present model with the results compared with those achieved using a front tracking method. Finally, a discussion on further work needed to extend the current model to model more complex solidification processes is provided.
Biographical Sketch

The author was born in Chongqing, P. R. China, 1978, the only child of Lida Wan and Xueqing Li. After she graduated from the first Chongqing High School, the author was admitted into Beijing University, Beijing in 1996, from where she received her Bachelor of Science (B.S.) degree in Mechanics and Engineering Science. She joined the Sibley School of Mechanical and Aerospace Engineering at Cornell University in the fall of 2000.
To my parents

Lida Wan and Xueqing Li
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Chapter 1

Introduction

1.1 Background

The understanding of metal alloy solidification processes is essential in many industrial applications, such as casting, welding, and growth of single crystals. Pure substances solidify with a sharp freezing front that coincides with the isotherm corresponding to the phase-change temperature \( T_f \). However, in multi-constituent systems the phase change takes place over a range of temperatures where solid and liquid may coexist in equilibrium. Although an alloy can solidify with a planar solid-liquid interface if the ratio of the freezing interface heat flux (G) and freezing interface growth velocity (v) is sufficiently large, most alloys solidify with the formation of a two phase region known as mushy zone, which is composed of solid dendrites and interdendritic liquid.

The shape, extent and advancement of the mushy zone depend on many factors including specific solidification conditions, initial composition of the liquid melt, etc. One of the most important factors is the convection in the melt and mushy region during solidification. There are numerous causes of fluid flow in casting processes.
The most common ones are thermal and solutal gradients; surface tension gradients, shrinkage and external forcing agents such as rotation, vibration, electromagnetic fields, etc. Natural convection can influence the advancement of the freezing front and local solidification rate even in highly conductive metals. Convection is also responsible for the segregation of alloy constituents during solidification, as well as affecting the size, orientation and structure of the resulting grains in the solidified bodies. Other defects in solidified alloys that are affected by convection include porosity and hot tears.

The nonuniform distribution of chemical composition and physical microstructure in an alloy can significantly affect mechanical properties of cast products. Hence, the design and control of the solidification process to achieve desired product with highly consistent properties have received considerable attention from many researchers in recent years. However, in order to control such processes, they must be well understood. The goal of the present study is to understand how the various convective transport phenomena affect macrosegregation patterns and freckle formation during solidification so that appropriate procedures can be employed to prevent macrosegregation. The investigation is done by mathematical modelling and numerical simulation.

Although laboratory experiments are valuable in understanding the connection between convection and macrosegregation, due to the high cost and the difficulty of flow visualization in opaque metals, available experiment data are very limited. Thus, mathematical modelling becomes more attractive for exploring novel processing schemes. Numerical simulation based on valid mathematical models offers opportunities to gain insights into various physical phenomena that are difficult, if not impossible, to extract through experiments.
Macrosegregation models are generally aimed at understanding the basic mechanisms involved, quantitatively predicting the occurrence and severity of macrosegregation, and performing studies for control and improvement of casting processes. The difficulty associated with the modelling of solidification processes arises from the morphological complexity and variety of length- and time-scales in the system. The geometry of the morphological structure between the solid/liquid interface in the mushy zone is very complicated and depends on the material properties and the phase change environment such as thermal conditions and species distribution. As an illustration, Figure 1.1 shows schematically two typical scales and corresponding physical phenomena that are involved in solidification. First, at the macroscopic scale, i.e. the scale of the casting system, fluid flow, convective-conductive heat transfer and macrosegregation and solid movement and deformation are present (Figure 1.1 a). Second, at the microscopic scale or morphological scale, interdendritic flow due to shrinkage, latent heat release due to phase change, nucleation and microstructure formation are present (Figure 1.1 b). Other scales also exist in the solidification process, such as the solute diffusion length scale and the capillary length scale [1, 2]. Furthermore, these macroscopic and microscopic phenomena are intimately coupled. For example, convection both affects and is affected by the microstructure of the dendrites within the mushy zone. Because of the multiple and often disparate length and time scales in a solidification system and the complex interaction between various physical phenomena, solving an entire solidification process as a microscopic problem is a formidable task. It requires grid resolutions of at least the dendritic arm spacing for numerical modelling so that the detailed morphologies can be captured. Such direct numerical simulation is impractical even with the computational power available today.
Figure 1.1: Different length-scales in a typical solidification process with a schematic of various convection patterns
A new approach to overcome these difficulties is to build and solve macroscopic and microscopic models separately and then couple them to simulate the interactions at different scales. In the macroscopic model, quantities like pressure and velocity of the fluid flow, temperature, concentration and volume fraction are the main variables and the purpose of the model is to predict the convection pattern, heat flux through the boundaries and macrosegregation. In the microscopic model, the focus is on the prediction of features such as microsegregation, grain size and orientation, columnar-to-equiaxed transition and porosity. The coupling is done by using the results obtained from the macroscopic model as known quantities in the microscopic model and vice-versa.

In the present study, a macroscopic mathematical model for binary alloy solidification processes is derived using a volume averaging technique. A finite element analysis is carried out for directional solidification of binary alloys. The effects of anisotropic permeability in the mushy zone are also investigated.

1.2 Related previous works

Modelling of macrosegregation has experienced explosive growth since the pioneering work of Flemings and coworkers in the mid-1960s [3, 4, 5]. By performing mass and species balances on the control volume element in the mushy zone, Flemings et al. derived the differential ‘Local Solute Redistribution Equation’ (LSRE). The model demonstrated how interdendritic flow is responsible for macrosegregation, but the analysis required prescription of the temperature and velocity field and the convection in the bulk liquid was ignored. Mehrabian et al. [6] proposed that the interdendritic flow velocity could be calculated from Darcy’s law for flow in porous media. Results obtained by this extended solute redistribution model for a
solidifying Al-4.5wt%Cu alloy compared favorably with experimental observations, but the temperature was taken from measurements. Kou et al. [7] used this model to predict the macrosegregation patterns for rotating, axisymmetric ingots. Fuji et al. [8] extended Mehrabian’s work to macrosegregation in multicomponent low-alloy steel. They refined the model by coupling the momentum and energy equations. A shortcoming of all of the above models is that they do not account for the coupling of fluid flow between the mushy and fully melted bulk liquid region.

Although greatly simplified, the early works of Szekely et al. [9] were significant because they marked the beginning of the development of sophisticated models for predicting transport phenomena during alloy solidification. Solid-liquid mixture equations were used in the mushy region and traditional single-phase equations were used in the melt. The first macrosegregation model that incorporated coupling of the flow between the mushy and bulk liquid region was reported by Ridder et al. [10] in 1981. They solved the coupled Darcy’s law, energy equation, and local solute redistribution equation in the mushy zone and the momentum and energy equations in the fully melted region. Solutal convection in the bulk liquid was neglected. In the numerical solution of the equations, a two-domain approach was employed in which the two sets of conservation equations are solved separately in mushy and all-liquid region and appropriate boundary conditions were explicitly satisfied at the solid/liquid interface. The iterative procedure involved matching pressure and velocity at the freezing front and remeshing was needed due to the advancement of the solidification front. Good agreement was obtained between the predicted and measured macrosegregation results.

The above models are multidomain in nature, because the mathematical solution domain is distinctly divided according to the physical domains (solid, mushy
and melt zones). Although this method gives good results for some examples, the usage is limited by the fact that the multidomain method requires the existence of discrete interfaces between regions or phases in the domain. The primary difficulty associated with the implementation of multidomain model centers on tracking the phase interface, which is generally an unknown function of space and time. The need for moving numerical grids and coordinate mapping procedures complicates the application of this technique and makes it impossible to predict highly irregular interface shapes that are present in most alloy systems.

Single-domain models, which overcome many of the limitations of the multidomain method, emerged in mid-1980s [11, 12] and later showed promise of becoming useful tools for simulating solidification processes. These models consist of a single set of equations for momentum, energy and species transport in multi-constituent, solid-liquid, phase change systems, which concurrently applied in all regions (solid, mushy and liquid). They require only a single, fixed numerical grid and a single set of boundary conditions to compute the solution. This brings a significant advantage in the numerical solution as the solid, mushy and liquid regions are implicitly coupled so there is no need to track the boundaries between phases. Instead, the interfaces are implicitly defined by the distributions of energy and composition determined from the solutions of the model equations (i.e. as a post-processing operation). The LSER of Flemings et al. can be derived from the single domain models as a limiting case. The momentum equations are a generalization of Darcy’ law and reduced to the single-phase Navier-Stokes equations in the liquid region.

The volume averaging model was developed independently by two groups of researchers. Bennon and Incropera derived a continuum model from classical mixture theory for binary, solid-liquid phase change systems [13, 14]. In their model, mushy
zone was viewed as solid-liquid mixture with macroscopic properties and limiting assumptions were invoked to reduce the number of dependent variables. In their paper, they also presented the first direct numerical simulation of freckles or A-segregation. Beckermann and Viskanta came up with a similar model in which the macroscopic transport equations are derived upon the classical microscopic transport equations using a volume averaging technique [15]. More recently, Prakash [16] and Ni and Beckermann [17] have presented models in which individual phase conservation equations are solved separately with the aid of interphase transport models. Such models permit the relaxation of assumptions pertaining to a non-deforming solid phase, thermal equilibrium ($T_s = T_l$) and negligible species diffusion through the solid phase, at the expense of solving twice as many partial differential equations.

1.3 Scope of study

It is well understood that the fluid flow in the liquid melt during solidification plays a very important role in macrosegregation in metallic alloys. In this thesis, a mathematical model to couple various important transport phenomena was derived using a volume averaging technique. This model belongs to the single-domain model family, requiring only a single fixed numerical grid and a single set of boundary conditions. The internal interfaces between liquid melt and mushy zone and between mushy and solid regions are implicitly defined by the distribution of temperature and volume fraction fields without the need to explicitly track the phase interfaces.

In terms of numerical implementation of the single domain model, almost all the reported works have been done by the finite difference method [14, 17, 18]. In the present work, the finite element method was used to solve the conservation equations. Three different finite element approaches are considered. The coupling
of the governing equations is achieved through an iterative procedure. In order to validate the numerical model and test its limitation, natural convection problems in porous media were investigated. The numerical results are compared with those of the earlier studies. Then the model was applied to directional binary alloy solidification in a rectangular domain. Comparison of the macrosegregation patterns with and without fluid flow showed that thermal-solutal convection is very important in determining macrosegregation under ground gravity level.

To further investigate the various physical phenomena occurring in solidification processes, the effects of anisotropic permeability of the dendritic mushy zone on macrosegregation pattern were studied. Pure aluminum solidification was also investigated as a limiting case using the single-domain model. Good agreements with result obtained by front-tracking method demonstrate the capability of the present numerical model. Finally, the limitation and possible extension of the present model in the simulation of directional alloy solidification are discussed.
Chapter 2

Volume-averaged macroscopic transport equations

2.1 Introduction

Physical phenomena in materials solidification can be described in detail by transport equations capable of handling the length scales that define the phase change at the level of the dendrites. However, the direct application of the transport equations to solidification modelling able to capture dendritic solidification is generally difficult, mainly because of the morphological complexities and the disparate time and length scales. So instead of solving the transport equations at the micro length scale, we develop a macroscopic model for the solidification process using a volume averaging approach.

The basic idea of volume averaging in order to obtain the macroscopic equations is to average the microscopic transport equations over a finite size control volume that contains both liquid and solid. The averaging volume as shown in Figure 2.1, is defined such that the scale it represents is small enough to capture the global fluid
flow motion, heat transfer, and species distribution as well as to track the liquid,
solid and mushy zone, but large enough to smooth out the details of the morpho-
logical complexities and the interdendritic fluid flow, latent heat release and species
redistribution. Under typical solidification conditions, the system and interfacial
structures are of the orders of $10^{-1}$ to $10^{0}$ m and $10^{-5}$ to $10^{-4}$ m respectively, so
the size of the averaging volume can vary between $10^{-3}$ to $10^{-2}$ m. The resulting
averaged transport equations need to be supplemented by constitutive relations that
describe morphological characteristics and the interactions between the two phases.

![Diagram of averaging volume containing columnar dendritic crystals](image)

Figure 2.1: Schematic illustration of the averaging volume containing columnar
dendritic crystals

It is possible to deduce the necessary terms in the macroscopic equations with-
out using an averaging process. For example, Hills et al. [12] and Bennon and
Incropera [13] utilized mixture theory to postulate macroscopic equations without
reference to any microscopic equations. However, the volume-averaging technique
provides many advantages [19]. Essentially, averaging shows how the various terms
in the macroscopic equations arise and how the resulting macroscopic variables are
related to the corresponding microscopic variables. This gives considerable insight
into the formulation of constitutive relations and holds the key for incorporating the evolution of the solid structure and the transport phenomena in the micro level into a macroscopic model. Therefore in this thesis, we have taken the volume averaging approach to derive a model for the columnar dendritic solidification of binary mixtures. In general, the averaging procedures and the form of the resulting equations are well established and have been utilized in the modelling of a large variety of multiphase systems [19, 20, 21]. For completeness, the definition of averaging quantities and related theorems are all included in this chapter.

2.2 Microscopic transport equations

According to Reynolds’ transport theorem, the general form of the microscopic transport equation of a certain quantity $\Phi$ can be rigorously derived from conservation laws and is given by:

$$\frac{\partial \Phi}{\partial t} + \nabla \cdot (\Phi \mathbf{v}) = \nabla \cdot \mathbf{J} + S$$  \hspace{1cm} (2.1)

where $\Phi$ can represent mass $\rho$, momentum $\rho \mathbf{v}$, enthalpy $\rho H$, or species concentration $\rho C$; $\mathbf{J}$ is a tensor of order one greater than that of $\Phi$ and accounts for the diffusion effect of momentum, heat or species; and $S$ is a source term.

The microscopic (exact) mass, momentum, energy and species transport equations for phase $k$ are given by:

\[
\begin{align*}
\text{Mass} & \quad \frac{\partial \rho_k}{\partial t} + \nabla \cdot (\rho_k \mathbf{v}_k) = 0 & (2.2) \\
\text{Momentum} & \quad \frac{\partial \rho_k \mathbf{v}_k}{\partial t} + \nabla \cdot (\rho_k \mathbf{v}_k \mathbf{v}_k) = \nabla \cdot \mathbf{\sigma}_k + \mathbf{b}_k & (2.3) \\
\text{Energy} & \quad \frac{\partial \rho_k h_k}{\partial t} + \nabla \cdot (\rho_k h_k \mathbf{v}_k) = -\nabla \cdot \mathbf{q}_k & (2.4) \\
\text{Species} & \quad \frac{\partial \rho_k C_k}{\partial t} + \nabla \cdot (\rho_k C_k \mathbf{v}_k) = -\nabla \cdot \mathbf{j}_k & (2.5)
\end{align*}
\]
where \( \sigma \) is the stress tensor, \( b \) is the body force, \( q \) is the heat flux, and \( j \) is the species diffusion flux. The detailed expressions for these terms are supplied by constitutive equations for specific cases.

The energy equation is written in terms of enthalpy. For simplicity, viscous heat dissipation, compression work, and volumetric energy and species sources are not included. Even though these assumptions are appropriate for most practical solidification systems, one can easily relax them as required.

### 2.3 Volume averaging approach

#### 2.3.1 Definitions and theorems

**Definition 2.3.1 phase function \( \nu_k \)**

The phase function \( \nu_k \) is a function of space and time, equalling 1 in phase \( k \) and zero elsewhere.

**Definition 2.3.2 volume fraction \( \epsilon_k \)**

The volume fraction of phase \( k \) is defined as

\[
\epsilon_k = \frac{1}{dV} \int_{dV} \nu_k(r,t) dv
\]

i.e. \( \epsilon_k = dV_k/dV \), where \( dV_k \) is the portion of \( dV \) that is occupied by phase \( k \), and \( dv \) is an infinitesimal element relative to \( dV \).

**Definition 2.3.3 volume-averaged quantity \( \langle \Psi_k \rangle \)**

The volume-average quantity is defined as

\[
\langle \Psi_k \rangle = \frac{1}{dV} \int_{dV} \Psi_k \nu_k(r,t) dv
\]
It is the averaged value of quantity $\Psi$ in phase $k$ over the entire averaging volume $dV$.

**Definition 2.3.4** *intrinsic volume-averaged quantity* $<\Psi_k>_k$

The intrinsic volume-average is defined as

$$<\Psi_k>_k = \frac{1}{dV_k} \int_{dV_k} \Psi_k \nu_k (r, t) dv$$  \hspace{1cm} (2.8)

The definition of $<\Psi_k>_k$ here represents the averaged value of $\Psi_k (r, t)$ in the control volume $dV_k$; if $\Psi_k (r, t)$ is uniformly distributed in $dV_k$, then $<\Psi_k>_k = \Psi_k (r, t)$. By comparing Equations (2.7) and (2.8), we obtain

$$<\Psi_k> = \epsilon_k <\Psi_k>_k$$  \hspace{1cm} (2.9)

with

$$\sum_k \epsilon_k = 1$$  \hspace{1cm} (2.10)

**Definition 2.3.5** *fluctuating component* $\hat{\Psi}_k$

The fluctuating component $\hat{\Psi}_k$ represents the deviation of $\Psi_k$ from the intrinsic volume average. It is given by

$$\hat{\Psi}_k = (\Psi_k - <\Psi_k>_k) \nu_k$$  \hspace{1cm} (2.11)

In phase $k$, it is zero only when $\Psi_k$ is uniformly distributed (i.e., $<\Psi_k>_k = \Psi_k$).

According to this definition, we have the following identity for changing the average of two products to the product of averaged quantities:

$$<\Psi_k \Phi_k> = \epsilon_k <\Psi_k>_k <\Phi_k>_k + <\hat{\Psi}_k \Phi_k>$$  \hspace{1cm} (2.12)

where

$$<\hat{\Psi}_k \Phi_k> = \frac{1}{dV} \int_{dV} \hat{\Psi}_k \Phi_k dv$$  \hspace{1cm} (2.13)
Gray [22] gives a very clear derivation of this identity.

The volume-averaging theorems listed below have been derived rigorously by several workers [22, 23, 24].

**Theorem 2.3.1** Relates the average of the time derivative to the time derivative of the average:

\[
< \frac{\partial \Psi_k}{\partial t} > = \frac{\partial < \Psi_k >}{\partial t} - \frac{1}{dV} \int_{dA_k} \Psi_k \mathbf{w}_k \cdot \mathbf{n}_k dA
\]  
(2.14)

where \(dA_k\) is the interfacial area of phase \(k\) with other phases, \(\mathbf{n}\) is the outward unit normal of the infinitesimal element of area \(dA\) of phase \(k\), and \(\mathbf{w}\) is the velocity of the microscopic interface.

**Theorem 2.3.2** Relates the average of the spatial derivative to the spatial derivative of the average:

\[
< \nabla \Psi_k > = \nabla < \Psi_k > + \frac{1}{dV} \int_{dA_k} \Psi_k \mathbf{n}_k dA
\]  
(2.15)

or

\[
< \nabla \Psi_k > = \epsilon_k \nabla < \Psi_k > + \frac{1}{dV} \int_{dA_k} \Psi_k \mathbf{n}_k dA
\]  
(2.16)

From a comparison of Equations (2.15) and (2.16), we also obtain that

\[
\frac{1}{dV} \int_{dA_k} < \Psi_k > \mathbf{n}_k dA = - < \Psi_k > \nabla \epsilon_k
\]  
(2.17)

and for \(\Psi_k = 1\), we conclude that:

\[
\frac{1}{dV} \int_{dA_k} \mathbf{n}_k dA = -\nabla \epsilon_k
\]  
(2.18)

**2.3.2 Derivation procedure**

The derivation summarized here basically follows most of the volume-averaging procedures reported in the literature [25, 24, 22]. Applying the general microscopic
transport equation (2.1) to phase \( k \), we obtain the following:

\[
\frac{\partial \Phi_k}{\partial t} + \nabla \cdot (\Phi_k \mathbf{v}) = \nabla \cdot \mathbf{J}_k + S_k \tag{2.19}
\]

Multiplying each side of Equation (2.19) by \( \nu_k \) and integrating it over the averaging volume \( dV \), we obtain

\[
\frac{1}{dV} \int dV \frac{\partial \Phi_k}{\partial t} \nu_k dV + \frac{1}{dV} \int dV \nabla \cdot (\Phi_k \mathbf{v}) \nu_k dV = \frac{1}{dV} \int dV \nabla \cdot \mathbf{J}_k \nu_k dV + \frac{1}{dV} \int dV S_k \nu_k dV \tag{2.20}
\]

Applying Theorem 1 to the first term we obtain

\[
\frac{1}{dV} \int dV \frac{\partial \Phi_k}{\partial t} \nu_k dV = \frac{\partial < \Phi_k >}{\partial t} + \frac{1}{dV} \int_{dA_k} \Phi_k \mathbf{w}_k \cdot \mathbf{n}_k dA \tag{2.21}
\]

Applying Theorem 2 to the second, third and fourth terms, results in the following:

\[
\frac{1}{dV} \int dV \nabla \cdot (\Phi_k \mathbf{v}) \nu_k dV = \nabla \cdot < \Phi_k \mathbf{v} > + \frac{1}{dV} \int_{dA_k} \Phi_k \mathbf{v} \cdot \mathbf{n}_k dA \tag{2.22}
\]

\[
\frac{1}{dV} \int dV \nabla \cdot \mathbf{J}_k \nu_k dV = \nabla \cdot < \mathbf{J}_k > + \frac{1}{dV} \int_{dA_k} \mathbf{J}_k \cdot \mathbf{n}_k dA \tag{2.23}
\]

\[
\frac{1}{dV} \int dV S_k \nu_k dV = < S_k > \tag{2.24}
\]

Furthermore, by applying the identity of Equation (2.12), \( \nabla \cdot < \Phi_k \mathbf{v}_k > \) in Equation (2.22) becomes

\[
\nabla \cdot < \Phi_k \mathbf{v}_k > = \nabla \cdot < \Phi_k > < \mathbf{v}_k > + \nabla \cdot < \Phi_k \mathbf{v}_k > \tag{2.25}
\]

Substituting Equations (2.21) - (2.24) into Equation (2.20), we obtain the following Equation:

\[
\frac{\partial < \Phi_k >}{\partial t} + \nabla \cdot < \Phi_k > < \mathbf{v}_k > = \nabla \cdot < \mathbf{J}_k > + < S_k > + \nabla \cdot \left( \frac{1}{dV} \int dV (\Phi_k \mathbf{v}_k) dV + \frac{1}{dV} \int_{dA_k} \mathbf{J}_k \cdot \mathbf{n}_k dA \right) + \frac{1}{dV} \int_{dA_k} (\Phi_k (\mathbf{w}_k - \mathbf{v}_k)) \cdot \mathbf{n}_k dA \tag{2.26}
\]
This is the exact form of the macroscopic transport equations. Compared with the exact microscopic equations (2.1), three extra terms appear from the volume averaging procedure. Let us define the following:

\[ I^D_k = \nabla \cdot \frac{1}{dV} \int_{dV} (-\Phi_k \vec{v}_k) dv \]  

(2.27)

\[ I^I_k = \frac{1}{dV} \int_{dA_k} \mathbf{J}_k \cdot \mathbf{n}_k dA \]  

(2.28)

\[ I^Q_k = \frac{1}{dV} \int_{dA_k} \Phi_k (\mathbf{w}_k - \mathbf{v}_k) \cdot \mathbf{n}_k dA \]  

(2.29)

We call \( I^D_k \) the microscopic deviation term and \( I^I_k \) and \( I^Q_k \) the interfacial terms, because \( I^D_k \) involves volume integration and \( I^I_k \) and \( I^Q_k \) involve surface integration. \( I^D_k \) appears because of the deviation of field quantities \( \Phi_k (\mathbf{r}, t) \) and \( \mathbf{v}_k (\mathbf{r}, t) \) from their intrinsically averaged counterparts.

From the definition of Equation (2.12), \( I^D_k \) is zero only when \( \Phi_k (\mathbf{r}, t) \) or \( \mathbf{v}_k (\mathbf{r}, t) \) is uniformly distributed in \( dV_k \), that is when \( \Phi_k \) or \( \mathbf{v}_k \) is zero. However, at the microscopic scale characterizing the mushy zone, there always exist species, temperature, and velocity gradients in the liquid, due to solute rejection, heat release and no-slip velocity boundary conditions; hence, \( \Phi_k (\mathbf{r}, t) \) and \( \mathbf{v}_k (\mathbf{r}, t) \) are usually not uniform within the averaging volume. Despite this fact, almost all solidification models reported in the literature neglect the microscopic deviation term [13, 24, 25], so we also neglect \( I^D_k \) in our derivation of the mathematical model. It can be seen from Equation (2.26) that the last two terms \( I^I_k \) and \( I^Q_k \) arise from the interfacial integration over \( dA \). \( I^Q_k \) accounts for the interfacial transfer due to phase change. Physically, it represents advection of interfacial mass, momentum, heat, or species of phase \( k \) due to the relative motion of the solid/liquid interface. \( I^I_k \) accounts for the effect of interfacial stress, heat and species transfer. It represents the transport phenomena between the phases within \( dV \) by diffusion and is related to the gradients of microscopic velocity, temperature, and species concentration on each side of
the solid/liquid interface $dA_k$ [17].

In order to solve Equation (2.26), macroscopic constitutive equations must be constructed to close the system. There are mainly two approaches in modeling these terms. The first one is called two-phase model, in which the transport equation (2.26) is applied to each phase and the resulting system of equations is solved separately [25]. The equations are coupled through the interfacial conditions estimated by $I^l_k$ and $I^Q_k$, which are expressed as a function of the interfacial area concentration $S_v$ and a mean interfacial flux. The main advantage of the two-phase model, as argued by Ni and Beckermann [17] is that it can account for non-equilibrium effects during solidification, such as rapid solidification, floating and settling of small equiaxed crystals, nucleation, etc. However, numerical computations with such a model require explicit treatment of the phase boundaries between highly irregular domains. In addition, a detailed and accurate formulation of the interfacial transfer terms has to be provided. The second approach is called one-phase model. In this model, the averaged macroscopic equations from different phases are added within the averaging volume $dV$, therefore, detailed modelling of the interfacial transfer terms $I^l_k$ and $I^Q_k$ can be avoided. The heat or mass lost from one phase is gained by other phases, i.e.

$$\sum_k I^l_k = 0 \quad \text{and} \quad \sum_k I^Q_k = 0 \quad (2.30)$$

Hence, the explicit treatment of interfacial flux is not necessary here. In the following section, the one-phase model will be presented, including the averaged macroscopic equations of mass, momentum, heat and species in the context of binary alloy solidification.
2.4 Mathematical model for columnar solidification of binary alloys

To make this model applicable to computation, the following basic assumptions are made.

**Assumption 2.4.1** Only solid and liquid phase may be present and no pores are formed, that is, $k = l$ or $s$ and $\epsilon_l + \epsilon_s = 1$.

**Assumption 2.4.2** Variations of material properties in $dV_k$ are neglected, although globally they may vary, that is

\[
< \rho_k >^k = \rho_k = < \rho_k >^l / \epsilon_k \quad \text{and} \quad < \mu_k >^k = \mu_k
\]

\[
< k_k >^k = k_k \quad \text{and} \quad < D_k >^k = D_k
\]

**Assumption 2.4.3** The flow is laminar and the solid phase is stationary, i.e. $< V_s >^s = < V_s >^l = V_s = 0$. This assumption is valid for columnar growth.

**Assumption 2.4.4** All phases in the averaging volume are in thermodynamic equilibrium, i.e. $< T_s >^s = < T_l >^l = T$.

**Assumption 2.4.5** There is no back diffusion in the solid phase, i.e. $D_s = 0$ and the liquid in the averaging volume is solutally well mixed, that is, $< C_l >^l = C_l$.

2.4.1 Macroscopic equation of mass conservation

In this case, $\Phi = \rho$, $J = 0$, and $S = 0$. According to Equation (2.26) the liquid and solid mass equations yield, respectively,

\[
\frac{\partial < \rho_l >}{\partial t} + \nabla \cdot (\epsilon_l < \rho_l >^l < v_l >^l) = I_l^Q
\]

\[
\frac{\partial < \rho_s >}{\partial t} + \nabla \cdot (\epsilon_s < \rho_s >^s < v_s >^s) = I_s^Q
\]
where

\[ I_l^Q = \frac{1}{dV} \int_{dV} (w_l - v_l) \cdot n_l dA \quad \text{and} \quad I_s^Q = \frac{1}{dV} \int_{dV} (w_s - v_s) \cdot n_s dA \] (2.33)

Here the microscopic deviation term does not appear due to Assumption 2.4.2. By adding Equation (2.31) and (2.32) and considering the interfacial mass balance, i.e. \( I_l^Q + I_s^Q = 0 \), and Assumption 2.4.2, we obtain:

\[ \frac{\partial (\epsilon_l \rho_l + \epsilon_s \rho_s)}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < v_l >^l + \epsilon_s \rho_s < v_s >^s) = 0 \] (2.34)

Furthermore, we define that

\[ \rho = \epsilon_l \rho_l + \epsilon_s \rho_s \] (2.35)

\[ v = \epsilon_l < v_l >^l \] (2.36)

According to Assumption 2.4.3, \( < v_s >^s = 0 \), the final form of the macroscopic mass conservation equation is

\[ \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho v) = 0 \] (2.37)

### 2.4.2 Macroscopic equation of momentum conservation

In this case \( \Phi = \rho v \) and \( S = b \) is the body force vector. Furthermore, we assume a Newtonian fluid and hence the viscous stress is terms of the rate of deformation is given as,

\[ J = -pI + \mu [\nabla v + (\nabla v)^T] \] (2.38)

The averaged momentum equations for the liquid and solid phases are given as follows:

\[ \frac{\partial \rho_l < v_l >}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < v_l >^l < v_l >^l) = -\nabla < p_l > + \nabla \cdot [< \mu_l \nabla v_l > + < \mu_l \nabla v_l >^T] + I_l^D + I_l^Q + I_l^I + < b_l > \] (2.39)
\[
\frac{\partial \rho_s < \mathbf{v}_s >}{\partial t} + \nabla \cdot (\epsilon_s \rho_s < \mathbf{v}_s >^s < \mathbf{v}_s >^s) = -\nabla < p_s > + \nabla \cdot [< \mu_s \nabla \mathbf{v}_s > + < \mu_s < \mathbf{v}_s >^T] + I_s^D + I_s^Q + I_s^I + < b_s > \quad (2.40)
\]

Following the argument about the deviation term, \( I_s^D = I_s^D = 0 \). According to Assumption 2.4.2, \( < \mu_k > = \mu_k \) and the above equations can be simplified as

\[
\frac{\partial (\rho_l < \mathbf{v}_l >)}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < \mathbf{v}_l >^l < \mathbf{v}_l >^l) = -\nabla < p_l > + \nabla \cdot (\epsilon_l \mu_l) [\nabla < \mathbf{v}_l >^l + < b_l >]
\]

\[
\frac{\partial (\rho_s < \mathbf{v}_s >)}{\partial t} + \nabla \cdot (\epsilon_s \rho_s < \mathbf{v}_s >^s < \mathbf{v}_s >^s) = -\nabla < p_s > + \nabla \cdot (\epsilon_s \mu_s) [\nabla < \mathbf{v}_s >^s + < b_s >]
\]

(2.41)

(2.42)

Let us add these two equations and notice that the interfacial momentum fluxes due to solidification balance each other, that is, \( I_l^Q + I_s^Q = 0 \). However, the interfacial stress in this case is not balanced; across the phase interface, there is a jump condition due to surface tension, \( I_l^I + I_s^I = \sigma \chi \), where \( \sigma \) is the surface tension, assumed to be constant, and \( \chi \) is the mean curvature of the interface.

Let us look carefully at the modelling of the \( I_l^I \) term. Flow through a mushy zone consisting of a continuous solid structure, such as columnar dendritic crystals, is usually very slow due to the high value of the interfacial area concentration. Therefore, the dissipative interfacial stress may be modelled in analogy with Darcy’s law as follows:

\[
I_l^I = -\frac{\epsilon_l \mu_l (< \mathbf{v}_l >^l - < \mathbf{v}_s >^s)}{K(\epsilon_l)}
\]

(2.43)

where \( K(\epsilon_l) \) is the permeability of the mushy zone. Values of the permeability have been reported for a columnar dendritic structure in the literature [26]. The permeability should approach infinity for \( \epsilon_l = 1 \) and zero for \( \epsilon_l = 0 \), which may be
accomplished by utilizing the Kozeny-Carman equation:

\[ K(\epsilon_l) = \frac{K_0 \epsilon_l^3}{(1 - \epsilon_l)^2} \tag{2.44} \]

Then the momentum equation in the liquid phase can be written as

\[
\frac{\partial (\rho_l < v_l >)}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < v_l > < v_l >) = -\nabla < p_l > + \nabla \cdot (\epsilon_l \mu_l)\nabla < v_l > + \nabla < v_l >^T] + I_l^Q - \frac{\epsilon_l \mu_l (< v_l >^l - < v_s >^s)}{K(\epsilon_l)} + < b_l > \tag{2.45}
\]

According to Assumption 2.4.3, \(< v_s >^s = 0\) and the definition of \(\rho\) and \(v\) (Equations (2.35) and (2.36)), the final form of the macroscopic transport equation of momentum conservation then yields

\[
\frac{\partial (\rho \mathbf{v})}{\partial t} + \nabla \cdot (\rho \mathbf{v}v) = -\nabla p + \mu \nabla \cdot [\nabla \mathbf{v} + (\nabla \mathbf{v})^T] - \frac{\mu}{K(\epsilon_l)} \mathbf{v} + \epsilon_l \rho \mathbf{g} \tag{2.46}
\]

where \(p = \epsilon_l < p_l >^l\), \(\mu = \epsilon_l \mu_l\), and \(\mathbf{g}\) is the unit gravity vector.

### 2.4.3 Macroscopic equation of energy conservation

In this case, \(\Phi = \rho h\), where \(h\) represents the total enthalpy and is defined as

\[
h = \int_0^T c dT + h_f \tag{2.47}
\]

and \(S = 0\). Furthermore, we utilize Fourier’s law for heat flux, that is, \(\mathbf{J} = -k \nabla T\). Other heat diffusion fluxes such as Soret and Dufour effects are neglected as they are less important in the convectional solidification process.

For the liquid and solid phases, the averaged energy equations are as follows:

\[
\frac{\partial (\rho_l < h_l >^l)}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < h_l >^l < v_l >^l) = \nabla \cdot < k_l \nabla T_l > + I_l^D + I_l^Q + I_l^J \tag{2.48}
\]

\[
\frac{\partial (\rho_s < h_s >^s)}{\partial t} + \nabla \cdot (\epsilon_s \rho_s < h_s >^s < v_s >^s) = \nabla \cdot < k_s \nabla T_s > + I_s^D + I_s^Q + I_s^J \tag{2.49}
\]
By applying Theorem 2.3.2 and using the Assumption 2.4.2, the first term on the right side of Equations (2.48) and (2.49) can be further expressed as:

\[
<k_k \nabla T_k> = \epsilon_k k_k \nabla <T_k>^k + k_k \frac{1}{dV} \int_{dA_k} \hat{T}_k n_k dA
\]  

(2.50)

where the term involving area integration is called the tortuosity factor [22] and is generally neglected in solidification processes.

Adding Equations (2.48) and (2.49) together, using the negligible deviation term assumption and the fact that the interfacial heat flux and heat transfer between phases are balanced with each other, that is,

\[
I_l^Q + I_s^Q = 0 \quad and \quad I_l^J + I_s^J = 0
\]  

(2.51)

we obtain:

\[
\frac{\partial (\rho_l < h_l >^l + \rho_s < h_s >^s)}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < h_l >^l < v_l >^l + \epsilon_s \rho_s < h_s >^s < v_s >^s) = \nabla \cdot [\epsilon_l k_l \nabla < T_l >^l + \epsilon_s k_s \nabla < T_s >^s]
\]  

(2.52)

By defining \(\rho h = \rho_l < h_l >^l + \rho_s < h_s >^s\) and \(k^* = \epsilon_l k_l + \epsilon_s k_s\) and by applying the Assumptions 2.4.3 and 2.4.4, the final form of the macroscopic transport equation of energy conservation then yields the following:

\[
\frac{\partial (\rho h)}{\partial t} + \nabla \cdot (\rho_l < h_l >^l v) = \nabla \cdot (k^* \nabla T)
\]  

(2.53)

### 2.4.4 Macroscopic equation of species conservation

In this case, \(\Phi = \rho C\), where \(C\) represents solutal concentration (per unit mass) and \(S = 0\). Furthermore, we utilize Fick’s law for species diffusion flux, that is, \(J = -\rho D \nabla C\).
Then for the liquid and solid phases, the averaged species equations yield, respectively, the following:

\[
\frac{\partial (\rho_l < C_l >^l)}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < C_l >^l < \mathbf{v}_l >^l) = \nabla \cdot (\rho_l D_l \nabla C_l) + I_l^D + I_l^Q + I_l^I \tag{2.54}
\]

\[
\frac{\partial (\rho_s < C_s >^s)}{\partial t} + \nabla \cdot (\epsilon_s \rho_s < C_s >^s < \mathbf{v}_s >^s) = \nabla \cdot (\rho_s D_s \nabla C_s) + I_s^D + I_s^Q + I_s^I \tag{2.55}
\]

Similar to the treatment of the energy equation, by applying Theorem 2.3.2 and using Assumption 2.4.2, the first term on the right side of Equations (2.54) and (2.55) can further be expressed as follows:

\[
< \rho_k D_k \nabla C_k > = \epsilon_k \rho_k D_k \nabla < C_k >^k + \rho_k D_k \frac{1}{dV} \int_{dA_k} \hat{C}_k \mathbf{n}_k dA \tag{2.56}
\]

where the second term on the right side of the equation is neglected in this solidification model.

Adding Equations (2.54) and (2.55) together, using the negligible deviation term assumption and the fact that the interfacial heat flux and heat transfer between phases are balanced with each other, that is,

\[
I_l^Q + I_s^Q = 0 \quad \text{and} \quad I_l^I + I_s^I = 0 \tag{2.57}
\]

we obtain the following energy equation:

\[
\frac{\partial (\rho_l < C_l >^l + \rho_s < C_s >^s)}{\partial t} + \nabla \cdot (\epsilon_l \rho_l < C_l >^l < \mathbf{v}_l >^l + \epsilon_s \rho_s < C_s >^s < \mathbf{v}_s >^s) = \nabla \cdot [\epsilon_l \rho_l D_l \nabla < C_l >^l + \epsilon_s \rho_s D_s \nabla < C_s >^s] \tag{2.58}
\]

By defining \( \rho C = \rho_l < C_l >^l + \rho_s < C_s >^s \) and \( D^* = \epsilon_l D_l \) and applying the Assumptions 2.4.3 and 2.4.4, the final form of the macroscopic transport equation of energy conservation can then be written as follows:

\[
\frac{\partial (\rho C)}{\partial t} + \nabla \cdot (\rho_l < C_l >^l \mathbf{v}) = \nabla \cdot (\rho_l D^* \nabla < C_l >^l) \tag{2.59}
\]
Chapter 3

Finite element analysis

3.1 Problem definition and solution strategy

Let us consider the directional solidification process of a dilute, incompressible binary alloy. The alloy is confined in mass impermeable mold walls. Initially the alloy melt is at a uniform temperature $T_i$ and concentration $C_i$. At time $t = 0^+$, a cooling heat flux is applied on the side $\Gamma^T$ of the mold wall to bring the boundary temperature below the eutectic temperature. The solidification commences from the side $\Gamma^T$ (see Figure 3.1).

Let us denote the solidification region by $\Omega \in \mathbb{R}^{n_{sd}}$, where $n_{sd}$ is the number of the space dimensions. The region $\Omega$ has a piecewise smooth boundary $\Gamma$ which consists of $\Gamma^T$ (boundary with prescribed temperature) and $\Gamma^q$ (boundary with prescribed heat flux).

Note that, because we adopt the single-domain model, there are no internal boundaries between the solid and mushy regions or the mushy zone and the liquid region. This is reflected on Figure 3.1, where no explicit interfaces are shown within the domain $\Omega$. 

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Figure 3.1: Schematic of the direct alloy solidification problem

The motion of the alloy melt is initially driven by the temperature gradient. As the solidification proceeds, solute is rejected at the solidification front due to different solubilities of the solid and liquid phases. Since the solute diffusivity is small, these rejected species pile up and create a concentration gradient in the mushy zone, which induces a solutal-driven fluid flow either enhancing or opposing the thermal-driven flow depending on the relative density of the rejected solute. If the rejected species is the lighter one of the two alloys, then the thermally driven and solutally driven flow are in the same direction and hence enhance each other; if instead the rejected solute is the heavier one, then the two flows will oppose each other.

The governing equations for the solidification system as derived in Chapter 2
are now introduced for the above defined problem. Let $\rho$ be the density, $k$ the conductivity, $c$ the heat capacity, $\mu$ the viscosity, $\alpha$ ($\alpha = k/\rho c$) the thermal diffusivity, $K_0$ the permeability and $D$ the solute diffusivity. The characteristic scale for time is taken as $L^2/\alpha$ and for velocity as $\alpha/L$. Hence, the dimensionless pressure is $p = \hat{p}L^2/\rho \alpha^2$. The dimensionless temperature $\theta$ is defined as $\theta \equiv (\hat{T} - T_e)/(T_0 - T_e)$ where $\hat{T}$, $T_0$ and $T_e$ are the real temperature, initial temperature and eutectic temperature, respectively. Likewise, the dimensionless concentration field $C$ is defined as $(\hat{C} - C_e)/(C_0 - C_e)$ where $\hat{C}$, $C_0$ and $C_e$ are the real concentration, initial concentration and eutectic concentration, respectively. The dimensionless enthalpy is defined as $h \equiv \hat{h}/c(T_0 - T_e)$. For simplicity, the subscript of liquid volume fraction is dropped throughout the following chapters, and the solid volume fraction $\epsilon_s$ is expressed as $1 - \epsilon$ with the help of the relation $\epsilon_l + \epsilon_s = 1$.

The key dimensionless quantities are the Prandtl number ($Pr = \nu/\alpha$), the Darcy number ($Da = K_0/L^2$), the Lewis number ($Le = \alpha/D$), the thermal Rayleigh number ($Ra_T = g\beta_T(T_0 - T_e)L^3/\nu \alpha$) and the solutal Rayleigh number ($Ra_C = g\beta_C(C_0 - C_e)L^3/\nu \alpha$). Here, $\beta_T$ and $\beta_c$ are the thermal and solutal expansion coefficients, respectively and $g$ is the gravity constant.

The other dimensionless parameters include the conductivity ratio $R_k$ ($k_s/k_l$), the heat capacity ratio $R_c$ ($c_s/c_l$) and the partition coefficient $\kappa$ (here the subscripts $s$ and $l$ are used to identify properties of the solid and liquid phases, respectively). The dimensionless slope $m$ of the liquidus line is given as $\hat{m}(C_0 - C_e)/(T_0 - T_e)$.

A no-slip condition on all boundaries $\Gamma$ provides a Dirichlet boundary condition for the fluid flow subproblem. It is assumed that adiabatic conditions are maintained on $\Gamma^v$. A problem dependent temperature condition on the boundary $\Gamma^T$ completes the definition of the thermal problem. The solutal boundary conditions are provided
by the impermeable wall condition on all boundaries. For completeness, the entire set of dimensionless coupled partial differential equations (PDEs) and boundary and initial conditions governing the solidification system are summarized in Table 3.1.

There are four conservation equations needed to be solved in this binary alloy solidification model. All of them are intimately coupled. However, the solution scheme implemented treats them in a decoupled manner, i.e. the equations are solved sequentially and iterations are performed to achieve convergence at each time step.

1. At time $t_n$, all fields such as velocity $v_n$, enthalpy $h_n$, temperature $\theta_n$, concentration $C_n$, liquid volume fraction $\epsilon_n$, etc. are known.

2. Advance to time step $t_{n+1} = t_n + \Delta t$. Set $i=0$, $v_{i=0}^{n+1} = v_n$, $h_{i=0}^{n+1} = h_n$, $\epsilon_{i=0}^{n+1} = \epsilon_n$, etc., where $i$ is an iteration index and the subscript denotes the time level.

3. Compute mixture enthalpy $h_{i+1}^{n+1}$ from energy equation 3.4.

4. Compute mixture concentration $C_{i+1}^{n+1}$ from solute equation 3.5.

5. Compute mixture velocity $v_{i+1}^{n+1}$ and pressure $p_{i+1}^{n+1}$ from the mass and momentum equations 3.1 and 3.2.

6. Update temperature $\theta_{i+1}^{n+1}$, liquid concentration $C_{i+1}^{n+1}$ and liquid volume fraction $\epsilon_{i+1}^{n+1}$ using a scheme that is described in section 3.4.

7. If $\|\theta_{i+1}^{n+1} - \theta_{i}^{n+1}\| < \epsilon_{tol}$, $\|C_{i+1}^{n+1} - C_{i}^{n+1}\| < \epsilon_{tol}$, $\|\epsilon_{i+1}^{n+1} - \epsilon_{i}^{n+1}\| < \epsilon_{tol}$ are all satisfied ($\epsilon_{tol}$ here denotes appropriate tolerance), then set $v_{n+1} = v_{i+1}^{n+1}$, $h_{n+1} = h_{i+1}^{n+1}$, $C_{n+1} = C_{i+1}^{n+1}$, etc.; set $n = n+1$ and proceed to step 2. Otherwise, set $i = i+1$ and proceed to step 3.
Table 3.1: Governing equations for binary alloy solidification problem.

\[ \nabla \cdot \mathbf{v}(x, t) = 0, \quad x \in \Omega \quad (3.1) \]

\[
\frac{\partial \mathbf{v}(x, t)}{\partial t} + \nabla \cdot \left( \frac{\mathbf{v}(x, t) \mathbf{v}(x, t)}{\epsilon} \right) = \nabla \cdot \mathbf{\sigma}(x, t) - \frac{(1 - \epsilon)^2}{\epsilon^3} \frac{Pr}{Da} \mathbf{v}(x, t) - \epsilon (Ra_T Pr \theta(x, t) - Ra_C Pr C_l(x, t)) \mathbf{e}_g, \quad x \in \Omega \quad (3.2) \]

\[ \mathbf{\sigma}(x, t) = -p(x, t) \mathbf{I} + Pr[\nabla \mathbf{v}(x, t) + (\nabla \mathbf{v}(x, t))^T], \quad x \in \Omega \quad (3.3) \]

\[
\frac{\partial h(x, t)}{\partial t} + \mathbf{v}(x, t) \cdot \nabla h_l(x, t) = \nabla \cdot [\epsilon + (1 - \epsilon)R_k] \nabla \theta(x, t)], \quad x \in \Omega \quad (3.4) \]

\[
\frac{\partial C(x, t)}{\partial t} + \mathbf{v}(x, t) \cdot \nabla C_l(x, t) = \frac{\epsilon}{Le} \nabla^2 C_l(x, t), \quad x \in \Omega \quad (3.5) \]

Initial conditions:

\[ \mathbf{v}(x, 0) = \mathbf{0}, \quad h(x, 0) = h_i, \quad C(x, 0) = C_i, \quad x \in \Omega \quad (3.6) \]

Boundary conditions:

\[ \mathbf{v}(x, t) = \mathbf{0}, \quad x \in \Gamma \quad (3.7) \]

\[ h(x, t) = h_s, \quad x \in \Gamma^T \quad (3.8) \]

\[ \frac{\partial h}{\partial n}(x, t) = 0, \quad x \in \Gamma^y \quad (3.9) \]

\[ \frac{\partial C}{\partial n}(x, t) = 0, \quad x \in \Gamma \quad (3.10) \]
3.2 Numerical scheme for the momentum equation

For the momentum equations, three types of finite element approaches have been implemented to obtain a solution:


2. The penalty function formulation, where the pressure is recovered a posteriori after the velocity field has been calculated satisfying the incompressibility condition approximately.

3. The fractional step method with mixed or equal order interpolation for velocity and pressure.

3.2.1 Stabilized finite element method

In the finite element implementation of the Navier-Stokes equations, stabilizing techniques are needed to enhance stability when equal-order-interpolation velocity-pressure elements are used. The most famous stabilization methods are the SUPG (Streamline-Upwind/Petrov-Galerkin) and the PSPG (Pressure-Stabilizing/Petrov-Galerkin) formulations proposed by many researchers (e.g. see Tezduyar et al. [27, 28]). Even though the SUPG/PSPG stabilizing method has been successfully used for incompressible flow problems, when it is applied to the momentum equation of a solidification system, it fails to yield the correct answers due to the presence of the Darcy term. Thus a new stabilizing term is needed for the generalized Darcy equation. This term is obtained by considering the Galerkin-Least-Squares for-
mulation of the steady-state equation. Let us denote with $R$ the residual of the steady-state momentum equation:

$$R = \nabla \cdot \left( \frac{vv}{\varepsilon} \right) - \nabla p + \frac{(1-\varepsilon)^2}{3} \frac{Pr}{Da} v + Pr \nabla^2 v - \varepsilon (Ra_T Pr \theta - Ra_C Pr C_l) e_g \quad (3.11)$$

By minimizing the sum of the squared residual integrated over each element domain i.e.

$$J[v, P] = \int_{\Omega} \frac{e^3}{(1-\varepsilon)^2} \frac{Pr}{Da} \left( \frac{1}{2} R^T R \right) d\Omega \quad (3.12)$$

we obtain $\delta J[v, P] = 0$ where $\delta J[v, P]$ is the following:

$$\delta J[v, P] = \int_{\Omega} \frac{e^3}{(1-\varepsilon)^2} \frac{Da}{Pr} R \left( \frac{1}{\varepsilon} v \cdot \nabla \delta v + \nabla \delta p - \frac{(1-\varepsilon)^2}{3} \frac{Pr}{Da} \delta v - Pr \nabla^2 \delta v \right) d\Omega \quad (3.13)$$

This procedure leads to a least square term with contributions from the advection, pressure, Darcy and the viscous stress term. We can write them explicitly as:

1. $\frac{\varepsilon}{\varepsilon} v \cdot \nabla w$ (advection term)
2. $c \nabla q$ (pressure term)
3. $c \frac{(1-\varepsilon)^2}{3} \frac{Pr}{Da} w = w$ (Darcy term)
4. $c Pr \nabla^2 w$ (viscous term)

where $w = \delta v$, $q = \delta P$ and $c = \frac{e^3}{(1-\varepsilon)^2} \frac{Da}{Pr}$. It is easy to identify that (1) gives the classical SUPG stabilizer and that (2) gives the PSPG stabilizer, respectively.

At the element interiors the contribution to the weighing function from the viscous term is identically zero for the P1P1 element and is neglected for the Q1Q1 element.

The third term, which we call DAPG, comes from the Darcy term and is the new stabilizer introduced for the momentum equation. As pointed out in [29], reversing the sign before the Darcy stabilizing term can help achieve better convergence rate, so we propose the form of the DAPG term as $-(1-\varepsilon)w$. 
Let us discretize the computation domain Ω by subdividing it into elements Ω^e, e = 1, 2, ..., n_{el}, where n_{el} is the number of elements. Associated with this discretization, we define the following finite element interpolation function spaces for the velocity, pressure, enthalpy and concentration:

\[ S_v^h = \{ v^h | v^h \in [H^1(Ω)]^{n_{el}}, v^h = 0 \; \text{on} \; Γ \}, \]

\[ V_w^h = \{ w^h | w^h \in [H^1(Ω)]^{n_{el}}, w^h = 0 \; \text{on} \; Γ \}, \]

\[ S_p^h = V_p^h = \{ q^h | q^h \in H^1(Ω) \}, \]

where \( S^h \) is the trial function space and \( V^h \) the test function spaces for the given finite element discretization. By adding the above three stabilizing terms, i.e. SUPG, PSPG and DAPG to the standard Galerkin formulation of the original time-dependent momentum equation, the stabilized Galerkin formulation of Equation 3.2 can be stated as follows: Find \( v^h \in S_v^h \) and \( p^h \in S_p^h \) such that, \( \forall \ w^h \in V_w^h \) and \( \forall \ q^h \in V_p^h \),

\[
\int_{Ω} w^h \cdot \left( \frac{∂v^h}{∂t} + \nabla \cdot (v^h v^h) \right) + \nabla \cdot (w^h) : \sigma(p^h, v^h) \, dΩ + \int_{Ω} q^h \nabla \cdot v^h \, dΩ \\
+ \sum_{e=1}^{n_{el}} \mathbf{A}_{e} \int_{Ω_e} (\delta^h + \epsilon^h + \gamma^h) \cdot \left[ \frac{∂v^h}{∂t} + \nabla \cdot (v^h v^h) \right] + (1 - \epsilon) \frac{1}{ρ} \mathbf{A}_{e} \varepsilon \cdot \sigma(p^h, v^h) - \epsilon (Ra_T Pr_θ - Ra_C Pr C_l) \mathbf{e}_g \right] \, dΩ = 0 \tag{3.14}
\]

The stabilizing terms \( δ^h \) and \( \epsilon^h \) in the fluid flow weak form are the SUPG and PSPG terms, respectively, whereas \( γ^h \) is the newly derived DAPG weighting function. The Petrov-Galerkin functions \( δ^h \), \( \epsilon^h \) and \( γ^h \) are defined as:

\[
δ^h = \frac{ε^3 Da}{(1 - ε)^2 Pr} τ_{SUPG} v^h \cdot \nabla w^h, \tag{3.15}
\]

\[
ε^h = \frac{ε^3 Da}{(1 - ε)^2 Pr} τ_{PSPG} \frac{1}{ρ} \nabla q^h, \tag{3.16}
\]
\[
\gamma^h = -(1 - \epsilon)w^h, \quad (3.17)
\]

where

\[
\tau_{SUPG} = \frac{h}{2 \| v^h \|} z(Rev), \quad (3.18)
\]

\[
\tau_{PSPG} = \frac{h^\#}{2 \| U \|} z(Re^\#).
\]

are the standard SUPG and PSPG stabilizing parameters. Notice that these two parameters are multiplied by a quantity \( \frac{\epsilon^2 Da}{(1 - \epsilon)^2 Pr} \) to obtain a smooth transition for the stabilizing terms across different regions in the solidification process. In the solid region where the fluid flow is absent, the SUPG and PSPG stabilizing terms will drop out as \( \epsilon \) approaches 0. When \( \epsilon \to 1 \), that is, in the liquid region, as the Darcy number \( Da \) is very small, effectively \( \frac{\epsilon^2 Da}{(1 - \epsilon)^2 Pr} \to 1 \), the modified stabilizing terms will be reduced to the standard SUPG and PSPG terms for pure fluid flow.

The spatial discretization of Equation (3.14) leads to the following set of nonlinear ordinary differential equations:

\[
\begin{align*}
&M + M_\delta + M_\gamma \{ \dot{v} \} + [N(v) + \ N_\delta(v) + \ N_\gamma(v)] \{ v \} \\
&\quad + [K + K_\delta + K_\gamma] \{ v \} + \ [D + D_\delta + D_\gamma] \{ v \} \\
&\quad - [G + G_\delta + G_\gamma] \{ p \} = \{ F(\theta, C_i) + F_\delta(\theta, C_i) + F_\gamma(\theta, C_i) \}, \quad (3.20)
\end{align*}
\]

\[
G^T \{ v \} + M_\epsilon \{ \dot{v} \} + N_\epsilon(v) \{ v \} + K_\epsilon \{ v \} \\
\quad + D_\epsilon \{ v \} + G_\epsilon \{ p \} = \{ F_\epsilon(\theta, C_i) \}, \quad (3.21)
\]

where \( \{ v \} \) is the vector of nodal values of \( v^h \), \( \{ \dot{v} \} \) is the time derivative of \( \{ v \} \), \( \{ p \} \) is the vector of nodal values of \( p^h \). The matrices \( M, N(v), K, D \) and \( G \) are derived, respectively, from the time-dependent, advective, viscous, Darcy and pressure terms. Note that only explicit dependencies on primary solution variables are shown. The matrix \( F \) is derived from the buoyancy term. The subscripts \( \delta \) and \( \epsilon \) identify the SUPG and PSPG contributions, respectively. The various matrices
forming the discrete finite element equations are described below, where the subscript \( p \) is introduced to denote the global velocity degrees of freedom in the final system of equations that corresponds to the \( i^{th} \) (\( i = 1, \ldots, n_{sd} \)) velocity degree of freedom of the elemental node \( \alpha \) in the \( e^{th} \) element. Similarly, one can define \( q \).

Here, \( P_e^e \) denotes the SUPG contribution, \( E_{\alpha i}^e \) denotes the PSPG contribution in the \( i^{th} \) direction and \( D_{\alpha i}^e \) denotes the DAPG contribution.

\[
\begin{align*}
[M + M_\delta + M_\gamma]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} \left( N_\alpha^e + P_\alpha^e + D_\alpha^e \right) N_{\beta,j}^e \Delta_{ij} \, d\Omega \right\} \quad (3.22) \\
[N(v) + N_\delta(v) + N_\gamma(v)]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} \frac{1}{\epsilon} \left( N_\alpha^e + P_\alpha^e + D_\alpha^e \right) N_{\beta,k}^e v_k \Delta_{ij} \, d\Omega \right\} \quad (3.23) \\
[K + K_\delta + K_\gamma]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} Pr \left( N_{\alpha,k}^e N_{\beta,k}^e + P_\alpha^e N_{\beta,kk}^e + D_\alpha^e N_{\beta,kk}^e \right) \Delta_{ij} \, d\Omega \right\} \quad (3.24) \\
[D + D_\delta + D_\gamma]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} \frac{(1 - \epsilon)^2}{\epsilon^3} Pr \left( N_\alpha^e + P_\alpha^e + D_\alpha^e \right) N_{\beta,ij}^e \Delta_{ij} \, d\Omega \right\} \quad (3.25) \\
[G + G_\delta + G_\gamma]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} \left( N_{\alpha,i}^e N_{eta,j}^e - P_{\alpha,i}^e N_{\beta,j}^e - D_{\alpha,i}^e N_{\beta,j}^e \right) \, d\Omega \right\} \quad (3.26) \\
[F + F_\delta + F_\gamma]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} \epsilon Pr \left( N_\alpha^e + P_\alpha^e + D_\alpha^e \right) \left( Ra_\lambda \theta_\beta^e - Ra_C \theta_\beta^e \right) e_{g_i} \, d\Omega \right\} \quad (3.27)
\end{align*}
\]

\[
\begin{align*}
[G^T]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} N_{\alpha,i}^e N_{\beta,j}^e \, d\Omega \right\} \quad (3.28) \\
[M_\epsilon]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} E_{\alpha,i}^e N_{\beta,j}^e \, d\Omega \right\} \quad (3.29) \\
[N_\epsilon(v)]_{pq} &= \frac{n_{el}}{\alpha} \left\{ \int_{\Omega_e} \frac{1}{\epsilon} E_{\alpha,i}^e v_k N_{\beta,k}^e \, d\Omega \right\} \quad (3.30)
\end{align*}
\]
The time integration of the Equations (3.20, 3.21) is performed using a backward Euler scheme, i.e.

\[
\dot{v} = \frac{v^n - v^{n-1}}{\Delta t}
\]  

(3.35)

The resulting nonlinear algebraic equations are solved by the Newton-Raphson method.

The advantage of the stabilizing method is that velocity and pressure are obtained simultaneously by solving one system of equations and the pressure field is usually smoother than that recovered a posteriori from the Poisson equation. However, the computation cost associated with solving the linear system using a direct solver is usually very expensive, and in some cases prohibitive. Thus, the Bi-conjugate gradient iterative method is used for the solution of the system to exploit the sparse nature of the matrix. Another difficulty with the stabilization method is that the matrix of the linear system is often ill-conditioned for very low Darcy number, which makes it practically impossible for such cases to find the correct solution. Although the stabilizing technique is successfully applied to the fluid flow in porous media problem, its application to solidification still can not be considered successful. More work needs to be done in this direction.
3.2.2 Penalty method

In the penalty formulation, the pressure is approximated by \( p = -\lambda \nabla \cdot v \) where, \( \lambda \) is the penalty number [30]. Then the continuity and momentum conservation are incorporated into one equation as:

\[
\frac{\partial v}{\partial t} + \nabla \cdot \left( \frac{vv}{\epsilon} \right) = \nabla (\lambda \nabla \cdot v) - \frac{(1 - \epsilon)^2 Pr}{\epsilon^3 Da} v - Pr \nabla \cdot [\nabla v + (\nabla v)^T] - \epsilon Pr (Ra_T \theta - Ra_C C_l) e_g \quad (3.36)
\]

Only the SUPG stabilizing term is needed here since the pressure has been eliminated from the momentum equation. The Galerkin formulation of Equation 3.36 can be written as follows: Find \( v^h \in S^h_v \) such that, \( \forall w^h \in V^h_v \),

\[
\int_{\Omega} w^h \cdot \left( \frac{\partial v^h}{\partial t} + \nabla \cdot \left( \frac{v^h v^h}{\epsilon} \right) - \nabla (\lambda \nabla \cdot v) + \frac{(1 - \epsilon)^2 Pr}{\epsilon^3 Da} v^h \right) - \epsilon Pr (Ra_T \theta - Ra_C C_l) e_g d\Omega + \int_{\Omega} \varepsilon (w^h) : (\nabla v + (\nabla v)^T) d\Omega
\]

\[
+ \sum_{e=1}^{n_{el}} A_e \int_{\Omega_e} \delta^h \cdot \left( \frac{\partial v^h}{\partial t} + \nabla \cdot \left( \frac{v^h v^h}{\epsilon} \right) - \nabla (\lambda \nabla \cdot v) + \frac{(1 - \epsilon)^2 Pr}{\epsilon^3 Da} v^h \right)
\]

\[- \nabla \cdot [\nabla v + (\nabla v)^T] - \epsilon Pr (Ra_T \theta - Ra_C C_l) e_g \right] d\Omega = 0 \quad (3.37)
\]

The semi-discrete form of Equation 3.37 is:

\[
[M + M_\delta] \{v\} + [N(v) + N_\delta(v)] \{v\} + [C + C_\delta] \{v\} + [K + K_\delta] \{v\} + [D + D_\delta] \{v\} = \{F(\theta, C_l) + F_\delta(\theta, C_l)\} \quad (3.38)
\]

The notation and significance of \( M, N(v), K, D \) and \( F \) are similar to those used for the stabilized FEM. The matrix \( C \) is derived from the penalty term contribution. The various matrices are described below:

\[
[M + M_\delta]_{pq} = \sum_{e=1}^{n_{el}} A_e \left( \int_{\Omega_e} (N^e_\alpha + P^e_\alpha) N^e_j \delta_{ij} d\Omega \right) \quad (3.39)
\]
\[
\begin{align*}
\left[ N(\mathbf{v}) + N_\delta(\mathbf{v}) \right]_{pq} &= \frac{n_{el}}{A} \left\{ \int_{\Omega_e} \frac{1}{\epsilon} (N^e_{\alpha} + P^e_{\alpha}) N^e_{\beta,j} v_k \Delta_{ij} \, d\Omega \right\} \quad (3.40) \\
\left[ C + C_\delta \right]_{pq} &= \frac{n_{el}}{A} \left\{ \int_{\Omega_e} \lambda (N^e_{\alpha,i} + P^e_{\alpha,j}) N^e_{\beta,j} \, d\Omega \right\} \quad (3.41) \\
\left[ K + K_\delta \right]_{pq} &= \frac{n_{el}}{A} \left\{ \int_{\Omega_e} P_{r}(N^e_{\alpha,k} N^e_{\beta,k} + P^e_{\alpha} N^e_{\beta,k,k}) \Delta_{ij} \, d\Omega \right\} \quad (3.42) \\
\left[ D + D_\delta \right]_{pq} &= \frac{n_{el}}{A} \left\{ \int_{\Omega_e} \frac{(1 - \epsilon)^2}{\epsilon^3} \frac{P_{r}}{D_{a}} (N^e_{\alpha} + P^e_{\alpha}) N^e_{\beta} \Delta_{ij} \, d\Omega \right\} \quad (3.43) \\
\left[ F + F_\delta \right]_{pq} &= \frac{n_{el}}{A} \left\{ \int_{\Omega_e} \epsilon P_{r} (N^e_{\alpha} + P^e_{\alpha})(Ra_\beta \theta^e_{\beta} - Ra_C C^e_{l,j}) e_{g,i} \, d\Omega \right\} \quad (3.44)
\end{align*}
\]

The Euler backward scheme is utilized for the time integration and the Newton-Raphson method is applied to solve the nonlinear equations. As pressure is eliminated from the momentum equation, the total degrees of freedom of the final linear system is greatly reduced, which makes the penalty method the most efficient one among the three methods that we have implemented. However, this advantage is at the same time a big drawback of the penalty formulation since the pressure field needs to be recovered from the obtained velocity field as a postprocessing operation.

### 3.2.3 Fractional step method

The fractional step method (also called the operator splitting method) was originally introduced by Chorin [31] in finite difference fluid flow applications. It was later extended to the finite element method by many authors. The velocity correction procedure used here is based on Patankar’s SIMPLER algorithm [32]. The algorithm includes two parts: the first part consists of the first two steps of the algorithm given below: calculation of the fictitious velocity, projection of the fictitious velocity into
a divergence-free subspace and approximate pressure field calculation; the second part consists of steps 3, 4 and 5 in the algorithm below: computing the velocity and pressure iteratively using the operator splitting scheme until convergence in $\mathbf{v}$ and $p$ is achieved. The overall algorithm is the following:

1. Calculate the fictitious velocity $\mathbf{v}^*$ by dropping the pressure term in the momentum equation. The nonlinear advection term is linearized by using the velocity from the previous time step. The diffusion and Darcy terms are treated implicitly.

$$\frac{\mathbf{v}^* - \mathbf{v}_n}{\Delta t} + \nabla \cdot \left( \frac{\mathbf{v}_n \mathbf{v}^*}{\epsilon} \right) = -\frac{(1 - \epsilon)^2}{\epsilon^3} \frac{Pr}{Da} \mathbf{v}^* + Pr \nabla \cdot [\nabla \mathbf{v}^* + (\nabla \mathbf{v}^*)^T]$$

$$- \epsilon Pr (RaT\theta_n - Ra_C C_{in}) \mathbf{e}_g \quad (3.45)$$

At this step, the velocity boundary condition is:

$$\mathbf{v}^* = 0 \quad \text{on} \quad \Gamma \quad (3.46)$$

2. Extract the pressure field from the fictitious velocity using the following Poisson equation:

$$\nabla^2 p_{n+1} = \frac{1}{\Delta t} \nabla \cdot \mathbf{v}^* \quad (3.47)$$

The pressure boundary condition at this step is:

$$\frac{\partial p_{n+1}}{\partial n} = 0 \quad \text{on} \quad \Gamma \quad (3.48)$$

$$p = 0 \quad \text{at node} \; 1 \quad (3.49)$$

3. Solve the momentum equation for velocity $\mathbf{v}_{n+1}^i$ using the newly obtained pressure field $p_{n+1}^i$:

$$\frac{\mathbf{v}_{n+1}^i - \mathbf{v}_n}{\Delta t} + \nabla \cdot \left( \frac{\mathbf{v}_n \mathbf{v}_{n+1}^i}{\epsilon} \right) = -\nabla p_{n+1}^i - \frac{(1 - \epsilon)^2}{\epsilon^3} \frac{Pr}{Da} \mathbf{v}_{n+1}^i$$

$$+ Pr \nabla \cdot [\nabla \mathbf{v}_{n+1}^i + (\nabla \mathbf{v}_{n+1}^i)^T] - \epsilon Pr (RaT\theta_n - Ra_C C_{in}) \mathbf{e}_g \quad (3.50)$$
4. Solve Poisson equation for the pressure-correction $\Delta p_i$ and correct pressure as

$$p_{n+1}^{i+1} = p_{n+1}^i + \Delta p_{n+1}^i$$

$$\nabla^2 (\Delta p_{n+1}^i) = \frac{1}{\Delta t} \nabla \cdot \mathbf{v}_{n+1}^i$$  \hspace{1cm} (3.51)

5. The fictitious velocities are corrected using the newly obtained pressure-correction $\Delta p_i$:

$$\mathbf{v}_{n+1}^{i+1} = \mathbf{v}_{n+1}^i - \nabla (\Delta p_{n+1}^i) \Delta t$$  \hspace{1cm} (3.52)

The boundary condition at this step is:

$$\mathbf{v}_{n+1}^{i+1} = 0 \quad \text{on} \quad \Gamma$$  \hspace{1cm} (3.53)

6. Check if $||\mathbf{v}_{n+1}^i - \mathbf{v}_{n}^i|| < \epsilon_{\text{tolerance}}$, go to next time step, otherwise return to step 3 and repeat until a converged solution is obtained.

As can be seen from the above description, Equation (3.45) for the fictitious velocity and Equation (3.50) for updating the velocity have almost identical forms. Similarly the forms of Equation (3.47) for approximate pressure and Equation (3.51) for pressure-correction are practically the same. Therefore in the following discussion, we focus on Equations (3.50) and (3.51).

The SUPG formulation for Equation (3.50) takes the following form:

$$\int_{\Omega} \mathbf{w}^h \cdot \left( \frac{\mathbf{v}_{n+1}^i - \mathbf{v}_n^i}{\Delta t} \right) + \nabla \cdot \left( \frac{\mathbf{v}_n^i \mathbf{v}_{n+1}^i}{\epsilon} \right) - \nabla p_{n+1}^i + \frac{(1-\epsilon)^2}{\epsilon^3} \frac{Pr}{Da} \mathbf{v}_{n+1}^i$$

$$- \epsilon Pr (Ra_T \theta - Ra_C C_l) \mathbf{e}_g d\Omega + \int_{\Omega} \varepsilon^2 (\nabla \mathbf{v}_{n+1}^i + (\nabla \mathbf{v}_{n+1}^i)^T) d\Omega \right) d\Omega$$

$$+ \sum_{e=1}^{n_{\text{el}}} \left[ \int_{\Omega_e} \delta_h \cdot [(\mathbf{v}_{n+1}^i - \mathbf{v}_n^i)/\Delta t] + \nabla \cdot (\mathbf{v}_n^i \mathbf{v}_{n+1}^i/\epsilon) - \nabla p_{n+1}^i + \frac{(1-\epsilon)^2}{\epsilon^3} \frac{Pr}{Da} \mathbf{v}_{n+1}^i$$

$$- \nabla \cdot (\nabla \mathbf{v}_{n+1}^i + (\nabla \mathbf{v}_{n+1}^i)^T) - \epsilon Pr (Ra_T \theta - Ra_C C_l) \mathbf{e}_g \right] d\Omega = 0$$  \hspace{1cm} (3.54)

The semi-discrete equation of this weak form is as follows:

$$[\mathbf{M} + \mathbf{M}_h] \{\dot{\mathbf{v}}\} + [\mathbf{N} - \mathbf{N}_h] \{\mathbf{v}\} + [\mathbf{G} + \mathbf{G}_h] \{\mathbf{v}\} + [\mathbf{K} + \mathbf{K}_h] \{\mathbf{p}\} = \mathbf{0}$$
\[ + [\mathbf{D} + \mathbf{D}_\delta] \{ \mathbf{v} \} = \{ \mathbf{F}(\theta, C_t) + \mathbf{F}_\delta(\theta, C_t) \} \]  

(3.55)

Since the matrices M, N(\mathbf{v}), K, D, and F are the same as those in the penalty formulation, their description is not repeated here.

The classical Galerkin formulation is applied to the pressure correction equation (3.51):

\[ \int_\Omega g^h (\nabla^2 (\Delta p_{n+1}^i) - \frac{1}{\Delta t} \nabla \cdot \mathbf{V}_{n+1}^i) \, d\Omega = 0 \]  

(3.56)

The matrix form of the Equation (3.56) is:

\[ [\mathbf{K}^p] \{ \Delta p_{n+1}^i \} = \frac{1}{\Delta t} [\mathbf{G}^T] \{ (\mathbf{v}_{n+1}^i)_{\beta k} \} \]  

(3.57)

where

\[ [\mathbf{K}^p]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} N_{\alpha,k}^e \, N_{\beta,k}^e \, d\Omega \right\} \]  

(3.58)

\[ [\mathbf{G}^T]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} N_{\alpha,k}^e \, N_{\beta}^e \, d\Omega \right\} \]  

(3.59)

The weak formulation for the velocity correction equation (3.52) is the following:

\[ \int_\Omega \mathbf{w}^h \cdot (\mathbf{v}_{n+1}^i + \mathbf{V}^i - \mathbf{v}^* + \nabla (\Delta p_{n+1}^i) \cdot \Delta t) \, d\Omega = 0 \]  

(3.60)

The matrix form of Equation (3.60) is

\[ [\mathbf{M}] \{ \mathbf{v}_{n+1}^i \} = [\mathbf{M}] \{ \mathbf{v}^* \} - \Delta t [\mathbf{G}] \{ \Delta p_{n+1}^i \} \]  

(3.61)

where

\[ [\mathbf{M}]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} N_{\alpha}^e \, N_{\beta,i}^e \, d\Omega \right\} \]  

\[ [\mathbf{G}]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} N_{\alpha}^e \, N_{\beta,i}^e \, d\Omega \right\} \]
Because the velocity and pressure are solved in a decoupled manner in the fractional step method, the demand on computational power is less intense than that of the stabilized FEM. In addition, due to its outstanding reliability, we chose this method to perform all the numerical simulations reported in this thesis.

### 3.3 Numerical scheme for the energy and species equations

Since both the energy and species conservation equations are scalar transport equations, a SUPG formulation with discontinuous weighing function is used in their implementation for stabilizing the corresponding advection terms. In order to facilitate computation, the energy and species equations are first cast into the standard form of a convection-diffusion scalar equation. For the energy equation, this involves rewriting the convection and diffusion parts in terms of the primary variable $h$. With the help of the identity

$$h_l = h + (1 - \epsilon)[(1 - R_c)\theta + h_f]$$

where $h_l$ is the liquid enthalpy and $h_f$ is the latent heat, and the approximation for the temperature gradient proposed by Amiez and Gremaud [33]

$$\nabla \theta^n \approx \alpha^{n-1} \nabla h^n$$

where $n$ denotes the time step and $\alpha^n$ is defined element wise by

$$\alpha^n = \begin{cases} \frac{\nabla \theta^n(x) \cdot \nabla h^n(x)}{||\nabla h^n(x)||^2} & \text{if } |\nabla h^n(x)| > 0, \\ \frac{\partial \theta}{\partial t}(x) & \text{if } |\nabla h^n(x)| = 0, \end{cases}$$

the energy equation can be written as

$$\frac{\partial h(x,t)}{\partial t} + \mathbf{v}(x,t) \cdot \nabla h(x,t) = -\nabla \cdot [(1 - \epsilon)((1 - R_c)\theta(x,t) + h_f)\mathbf{v}(x,t)]$$
The weak formulation of the energy equation (3.64) can be stated as follows:

Find \( h^h \in S^h_\theta \) such that, \( \forall h^h \in V^h_\theta \):

\[
\int_\Omega \hat{w}^h \left( \frac{\partial h^h}{\partial t} + \bm{v}^h \cdot \nabla h^h \right) d\Omega + \int_\Omega \nabla \hat{w}^h \cdot \left( (\epsilon + (1 - \epsilon) R_k^e) \alpha \nabla h^h \right) d\Omega \]

\[+ \sum_{e=1}^{n_{el}} \int_{\Omega_e} \hat{\delta}^h \left( \frac{\partial h^h}{\partial t} + \bm{v}^h \cdot \nabla h^h \right) d\Omega_e \]

\[= - \int_\Omega (\hat{w}^h + \hat{\delta}^h) \left( \bm{v}^h \cdot \nabla [(1 - \epsilon)((1 - R_c)^h + h_f^h)] \right) d\Omega \quad (3.65)\]

The spatial discretization of equation (3.65) leads to the following ordinary differential equation:

\[ [\hat{M} + \hat{M}_\delta] \{ \dot{h} \} + [\hat{N} + \hat{N}_\delta] \{ \dot{h} \} + [\hat{K} + \hat{K}_\delta] \{ h \} = \{ \hat{F} + \hat{F}_\delta \} \quad (3.66) \]

The element stiffness matrices and the load vector are given as follows:

\[
[\hat{M} + \hat{M}_\delta]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} (N^e_\alpha + \delta^e_\alpha) N^e_\beta d\Omega \right\} \quad (3.67)
\]

\[
[\hat{N} + \hat{N}_\delta]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} (N^e_\alpha + \delta^e_\alpha) v^p_k N^e_\beta d\Omega \right\} \quad (3.68)
\]

\[
[\hat{K} + \hat{K}_\delta]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} (\epsilon + (1 - \epsilon) R_k^e) \alpha \ N^e_{a,k} N^e_{b,k} d\Omega \right\} \quad (3.69)
\]

\[
[\hat{F} + \hat{F}_\delta]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} (N^e_\alpha + \delta^e_\alpha)(1 - \epsilon) R_c^e \theta_{i,j} v^p_k - v^p_k \epsilon_{i,j} ((1 - R_C^e) \theta + h_f) ) d\Omega \right\} \quad (3.70)
\]

Based on the same argument, we rewrite the species conservation equation as:

\[
\frac{\partial C(x,t)}{\partial t} + \bm{v}(x,t) \cdot \nabla C(x,t) = \frac{\epsilon}{L_c} \nabla^2 C(x,t) - C_i(x,t) + \frac{\epsilon}{L_e} \nabla^2 (C_i(x,t) - C(x,t)) \quad (3.71)
\]
There are various advantages in writing the species equation in this form. At first, the SUPG algorithm can be easily applied by dumping the effect of the liquid concentration $C_l$ into a body force term. Secondly, the above form allows implementation of different microscopic diffusion models without changing the form of the governing equation.

The weak formulation of the species equation (3.71) can be stated as follows:

Find $C^h \in S^h_C$ such that, $\forall \; \tilde{w}^h \in V^h_C$:

$$
\int_{\Omega} \tilde{w}^h \left( \frac{\partial C^h}{\partial t} + \mathbf{v}^h \cdot \nabla C^h \right) d\Omega + \int_{\Omega} \nabla \tilde{w}^h \cdot \left( \frac{\epsilon}{Le} \nabla C^h \right) d\Omega + \sum_{e=1}^{n_{el}} \mathbf{A} \int_{\Omega_e} \tilde{\delta}^h \left( \frac{\partial C^h}{\partial t} + \mathbf{v}^h \cdot \nabla C^h \right) d\Omega_e
= \int_{\Omega} \left( \tilde{w}^h + \tilde{\delta}^h \right) \left( (\mathbf{v}^h \cdot \nabla (C^h - C_l^h)) + \frac{\epsilon}{Le} \nabla^2 (C^h_l - C^h) \right) d\Omega
$$

(3.72)

The spatial discretization of equation (3.72) leads to the following ordinary differential equation:

$$
[\tilde{\mathbf{M}} + \tilde{\mathbf{M}}_\delta] \{\dot{C}\} + [\tilde{\mathbf{N}} + \tilde{\mathbf{N}}_\delta] \{C\} + [\tilde{\mathbf{K}} + \tilde{\mathbf{K}}_\delta] \{C\} = \{\tilde{\mathbf{F}} + \tilde{\mathbf{F}}_\delta\}
$$

(3.73)

The element stiffness matrices and the load vector are given as follows:

$$
[\tilde{\mathbf{M}} + \tilde{\mathbf{M}}_\delta]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} (N^e_{\alpha} + \delta^e_{\alpha}) N^e_{\beta} d\Omega \right\}
$$

(3.74)

$$
[\tilde{\mathbf{N}}(\mathbf{v}) + \tilde{\mathbf{N}}_\delta(\mathbf{v})]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} (N^e_{\alpha} + \delta^e_{\alpha}) v^e_k N^e_{\beta,k} d\Omega \right\}
$$

(3.75)

$$
[\tilde{\mathbf{K}} + \tilde{\mathbf{K}}_\delta]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} \frac{\epsilon}{Le} N^e_{\alpha,k} N^e_{\beta,k} d\Omega \right\}
$$

(3.76)

$$
[\tilde{\mathbf{F}} + \tilde{\mathbf{F}}_\delta]_{pq} = \sum_{e=1}^{n_{el}} \mathbf{A} \left\{ \int_{\Omega_e} \left( N^e_{\alpha} + \delta^e_{\alpha} \right) \left[ (C_{,k} - C_{l,k}) v_k + \frac{\epsilon}{Le} (C_{l,k,k} - C_{l,k}) \right] d\Omega \right\}
$$

(3.77)
The time integration of Equations (3.66) and (3.73) is achieved by using the standard Newmark scheme [30, 34].

### 3.4 Supplementary relationships

In the above derived single-domain model, we have four governing equations but seven unknown variables: velocity $v$, pressure $p$, mixture enthalpy $h$, mixture concentration $C$, temperature $\theta$, liquid volume fraction $\epsilon$, and liquid concentration $C_l$. To close the model, we need three supplementary relationships for $\theta$, $\epsilon$, and $C_l$.

**Enthalpy-temperature relation.** Enthalpy and temperature are related by the specific heat. By assuming that all phases in the averaging volume are in thermal equilibrium, that is, $\theta_l = \theta_s = \theta$, the general expression for the mixture enthalpy is:

$$h = (\epsilon + (1 - \epsilon)R) \theta + \epsilon \ h_f + \frac{R_c T_e}{\Delta T}$$

where the last term in the equation comes from the non-dimensionalization process.

**Thermodynamic relations.** For a wide range of practical applications, solidification of binary systems can be closely approximated by the assumption that the liquid in the averaged volume is solutally well mixed, that is, $C_l = C_{\text{liquidus}}$, and at the liquid-solid interface, the relation of solidus and liquidus concentration is given by the phase diagram. In reference to the binary phase diagram of Figure 3.2 with straight liquidus and solidus lines, local conservation of concentration can be expressed using the lever rule:

$$C = \epsilon \ C_l + (1 - \epsilon) \ C_s$$

With the definition of the equilibrium partition ratio $k_p = C_s/C_l$, a direct relationship between the mixture concentration $C$ and liquid concentration $C_l$ is obtained.
The liquidus concentration and the temperature are related by:

\[ C_{\text{Liquidus}} = \frac{\theta - \theta_m}{\theta_e - \theta_m} C_e \]  
(3.80)

By using the above three supplementary relationships, we propose an update scheme for temperature \( \theta \), liquid concentration \( C_l \) and liquid volume fraction \( \epsilon \) as follows: First, we define some quantities according to our analysis:

\[ \theta_{\text{Liquidus}} = \theta_m + m C \]  
(3.81)

\[ T_{\text{Solidus}} = \max\{\theta_m + \frac{m}{k} C, \theta_e\} \]  
(3.82)

\[ h_{\text{Liquidus}} = \theta_{\text{Liquidus}} + (R_e - 1)\theta_e + h_f \]  
(3.83)

\[ h_{\text{Solidus}} = R_e \theta_{\text{Solidus}} \]  
(3.84)

\[ h_e = h_{\text{Solidus}} + \left[1 - \frac{1}{1 - k_p} (\frac{\theta_{\text{Solidus}} - \theta_{\text{Liquidus}}}{\theta_{\text{Solidus}} - \theta_m})\right] h_f \]  
(3.85)
With the definition of $h_{\text{Liquidus}}$, $h_{\text{Solidus}}$, and $h_e$, the phase diagram is divided into four different regions according to the relationships between the mixture enthalpy and the three critical enthalpy values:

- When $h > h_{\text{Liquidus}}$, the control volume is occupied by pure liquid, corresponding to region (1) in the phase diagram.

- When $h_e < h < h_{\text{Liquidus}}$, solidification has started but the liquid concentration has not reached the eutectic point, corresponding to region (2) in the phase diagram.

- When $h_{\text{Solidus}} < h < h_e$, the melt in the control volume is solidified at eutectic temperature and eutectic concentration, corresponding to region (3) in the phase diagram.

- When $h < h_{\text{Solidus}}$, the alloy in the averaging volume is completely solidified, corresponding to region (4) in the phase diagram.

One we calculate $h_{\text{Liquidus}}$, $h_{\text{Solidus}}$, and $h_e$ corresponding to a certain mixture concentration $C$, we can compare the value of mixture enthalpy $h$ obtained from the energy equation to them to determine which area of the phase diagram the node is in. Then the following scheme is utilized to update the values of temperature, liquid concentration and liquid volume fraction:

- When $h <= h_{\text{Solidus}}$, $\theta = \frac{h}{R_c}$, $\epsilon = 0$ and $C_l = C_e$,

- When $h_{\text{Solidus}} < h <= h_e$, $\theta = \theta_e$, $\epsilon = 1 - \frac{h-h_{\text{Solidus}}}{h_f}$ and $C_l = C_e$,

- When $h_e < h <= h_{\text{Liquidus}}$, $\theta = \frac{h-e((R_e-1)\theta_e+h_f)}{e(1-e)\theta_e}$, $\epsilon = [1 - \frac{1}{1-k_p}(\frac{\theta-\theta_{\text{Liquidus}}}{\theta_{\text{m}}})]$ and $C_l = \frac{C}{1+\epsilon(k_p-1)}$

- When $h_{\text{Liquidus}} < h$, $\theta = h - ((R_c-1)\theta_e + h_f)$, $\epsilon = 1$ and $C_l = C$
Chapter 4

Numerical examples

4.1 Fluid flow in porous media

In the process of deriving the volume-averaged macroscopic equations for the binary alloy system, the mushy zone is treated as a porous media with solid dendrites and interdendritic liquid. To test the applicability of the macroscopic transport equations to the solid/liquid two-phase system, a series of fluid flow problems in porous media are investigated using the present model. In the absence of phase change, liquid volume fraction is constant during the simulation, which gives a linear relationship between enthalpy and temperature and between the mixture concentration and the liquid concentration. Therefore the supplementary relationships are simplified to two linear equations. All the example problems in this section have been taken from [35, 36].

4.1.1 Example 1

Let us consider a two-dimensional square cavity consisting of a solid matrix and saturated fluid with uniform liquid volume fraction of 0.8. The system is initially
Figure 4.1: Geometry and boundary conditions of the natural convection problem with uniform porosity

at a dimensionless homogeneous temperature $\theta_0 = 1.0$. The top and bottom walls of the cavity (see Figure 4.1) are insulated. The right wall ($x = 1.0$) is maintained at the initial temperature for times $t > 0$ while a constant temperature of $\theta = 1$ is prescribed at the left wall ($x = 0.0$). No-slip velocity boundary conditions are applied to all walls. The governing dimensionless physical parameters are Prandtl number 1.0, Darcy number $10^{-6}$, and thermal Rayleigh number $10^8$. A slightly biased mesh of $(36 \times 36)$ (see Figure 4.2) and dimensionless time step $\Delta t = 0.0001$ are used for the computation. The predicted streamline and isothermal patterns are similar to those of natural convection in pure liquid except that the magnitude of velocity is much smaller in the porous media case. The results agree very well with those in [36].
4.1.2 Example 2

As both heat and mass transfer are present in solidification of binary alloys, a double diffusive convection problem in uniform porosity media is investigated to demonstrate the validity of the macroscopic transport equations. A square cavity with adiabatic horizontal walls and vertical walls with prescribed temperature is considered. All properties except density are assumed to be constant, and density variation is represented by the Boussinesq approximation. The porosity is uniform and taken as 0.6 throughout the cavity. The dimensionless initial temperature $\theta_0$ and concentration $C_0$ both equal to 0. The boundary conditions are shown in Figure 4.4. A finite element grid of $(50 \times 50)$ is used and the mesh is slightly biased toward the wall to resolve the boundary layers. The system is characterized by the dimensionless parameters: Prandtl number 1.0, Lewis number 2, thermal Rayleigh
Figure 4.3: Streamline and isothermal patterns of natural convection in a porous material with uniform porosity
number $2 \times 10^8$ and solutal Rayleigh number $-1.8 \times 10^8$. The steady-state solution is achieved at dimensionless time step $t = 1.2$ and the streamline, isothermal, and isoconcentration patterns (Figures 4.6 and 4.7) are in excellent agreement with the results obtained in [35].

4.1.3 Example 3

In the binary alloy solidification process, the porosity of the dendritic matrix varies from the tip of the dendrites to the eutectic front, thus the study of the convection in variable porosity media is helpful in validating the applicability of the volume-averaged transport equations to the solidification system. This example problem is similar to the system studied in Example 1. In particular, the same geometry, boundary conditions are considered as in Example 1. However the physical parameters are different. The porosity varies linearly with distance from the walls to the center (see Figure 4.8). The wall porosity $\epsilon_w$ is 0.4, the thickness of the porous
Figure 4.5: Computational mesh of the double diffusive convection problem with uniform porosity media is 30% of the length of the square cavity. The Darcy number $Da$ and thermal Rayleigh number $Ra_T$ are $6.665 \times 10^{-7}$ and $10^6$, respectively. The computational grid is the same as that used in Example 1. Predicted streamline and isothermal patterns are presented in Figures 4.9. Unlike the case of uniform porosity, a twin vortex flow pattern is observed for the variable porosity media. The counterclockwise circulation near the hot wall is induced by the high velocity region which occurs at the interface between the porous medium layer and the whole liquid region. It is interesting to note that even at high Rayleigh numbers, the conduction effects are still predominant near the walls, while distortion of isotherms due to convection is observed in the core of the cavity where the solid matrix is absent ($\epsilon = 1.0$).
4.2 Directional binary alloy solidification

4.2.1 Example 1

Binary alloy solidification is considered in a two-dimensional rectangular impermeable cavity of dimensions 25mm × 100mm as shown schematically in Figure 4.10. Initially, all boundaries are insulated, the cavity is charged with a superheated binary solution of composition 0.7 and temperature 311 K and the system is in equilibrium. At time $t = 0^+$, the temperature of the left wall is suddenly reduced to and maintained at 223 K, which is less than the eutectic temperature 257.75 K, while the right wall is maintained at the initial temperature. Calculation is performed for an aqueous solution of ammonium chloride ($NH_4Cl - H_2O$). This semi-transparent alloy system has been selected in many studies mainly because it permits flow visu-
alization. The thermophysical properties of this system are listed in Table 4.1. The resulting dimensionless quantities are listed in Table 4.2. A finite element mesh of $41 \times 41$ bilinear elements was used for computation and the grid is slightly biased toward the walls to resolve the boundary layers (see Figure 4.11).

In this example, we study the phase change phenomena in the absence of convection, i.e. the momentum equation is left out and only energy and solute transport equations are solved. The isothermal and isoconcentration patterns are plotted in Figure 4.12 - 4.13. As there is no fluid flow, solidification is a pure diffusion process, hence both liquidus and solidus fronts propagate without vertical temperature and composition gradients. Significant temperature gradients are confined to the solid and mushy regions. Similarly, liquid composition gradients are confined primarily to the mushy regions, and the bulk fluid remains very nearly at the initial composition. Such conduction dominated solidification process can be achieved under reduced gravity levels where the buoyancy force is negligible.

### 4.2.2 Example 2

Let us consider the same solidification problem as in Example 1 except that in this example the thermal-solutal convection is taken into consideration. At $t = 0^+$ the freezing process starts and takes place under standard laboratory conditions. Due to the rapid transient associated with solidification simulations at early times, a small dimensionless time step of $\Delta \tau = 2 \times 10^{-5}$ was utilized for the first 250 steps, after
### Table 4.1: Physical constants for the NH$_4$Cl - 30\% H$_2$O system

<table>
<thead>
<tr>
<th>Property</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Density of the solid</td>
<td>$\rho_s (kg/m^3)$</td>
<td>1078</td>
</tr>
<tr>
<td>Density of the liquid</td>
<td>$\rho_l (kg/m^3)$</td>
<td>1078</td>
</tr>
<tr>
<td>Thermal conductivity of the solid</td>
<td>$k_s (W/mK)$</td>
<td>0.393</td>
</tr>
<tr>
<td>Thermal conductivity of the liquid</td>
<td>$k_l (W/mK)$</td>
<td>0.468</td>
</tr>
<tr>
<td>Solid specific heat</td>
<td>$c_s (J/kgK)$</td>
<td>1870</td>
</tr>
<tr>
<td>Liquid specific heat</td>
<td>$c_l (J/kgK)$</td>
<td>3249</td>
</tr>
<tr>
<td>Diffusion coefficient</td>
<td>$D (m^2/s)$</td>
<td>$4.8 \times 10^{-9}$</td>
</tr>
<tr>
<td>Viscosity of the liquid</td>
<td>$\mu (kg/m/s)$</td>
<td>$1.3 \times 10^{-3}$</td>
</tr>
<tr>
<td>Latent heat</td>
<td>$h_f (J/kg)$</td>
<td>$3.138 \times 10^5$</td>
</tr>
<tr>
<td>Permeability coefficient</td>
<td>$K_0 (m^2)$</td>
<td>$5.56 \times 10^{-11}$</td>
</tr>
<tr>
<td>Thermal expansion coefficient</td>
<td>$\beta_T (K^{-1})$</td>
<td>$3.832 \times 10^{-4}$</td>
</tr>
<tr>
<td>Solutal expansion coefficient</td>
<td>$\beta_c$</td>
<td>0.257</td>
</tr>
<tr>
<td>Eutectic temperature</td>
<td>$T_e (K)$</td>
<td>257.75</td>
</tr>
<tr>
<td>Eutectic composition</td>
<td>$C_e$</td>
<td>0.803</td>
</tr>
<tr>
<td>$NH_4Cl - H_2O$ melting point</td>
<td>$T_m (K)$</td>
<td>633.59</td>
</tr>
<tr>
<td>Equilibrium partition ratio</td>
<td>$\kappa_p$</td>
<td>0.30</td>
</tr>
</tbody>
</table>
Table 4.2: Dimensionless groups and their characteristic values

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Prandtl number</td>
<td>Pr 9.025</td>
</tr>
<tr>
<td>Lewis number</td>
<td>Le 27.84</td>
</tr>
<tr>
<td>Darcy number</td>
<td>Da $8.896 \times 10^{-8}$</td>
</tr>
<tr>
<td>Thermal Rayleigh number</td>
<td>$Ra_T 1.938 \times 10^6$</td>
</tr>
<tr>
<td>Solutal Rayleigh number</td>
<td>$Ra_c -2.154 \times 10^6$</td>
</tr>
<tr>
<td>Dimensionless slope of</td>
<td>$m 0.905$</td>
</tr>
<tr>
<td>the liquidus</td>
<td></td>
</tr>
<tr>
<td>Heat conductivity ratio</td>
<td>$R_k 0.840$</td>
</tr>
<tr>
<td>Heat capacity ratio</td>
<td>$R_c 0.576$</td>
</tr>
</tbody>
</table>

which the time step was increased to $5 \times 10^{-5}$ for 900 steps and to 0.0001 for all the rest solidification process, as the solidification proceeds. The system achieved steady-state at dimensionless time $\tau = 0.14$.

Figures 4.16 - 4.19 illustrate the velocity, streamlines, isotherms and liquid iso-concentration distributions at dimensionless times of $\tau = 0.009, 0.018, 0.07$ and 0.14, respectively. Velocity vectors shown in each figure are based on the mixture velocity components $u = \epsilon_l u_l$ and $v = \epsilon_l v_l$. As can be seen from the plots, conditions are conduction dominated at the beginning of the process, and the simulation showed the transient development of convection patterns. Convection in the melt was thermally driven by heating from the hot right wall and cooling through the solid and mushy zones, and solutally driven flow was predicted in the mushy zone. The thermal and solutal buoyancy forces are opposing each other due to the fact that water, the lighter solute of the two, was rejected into the liquid. The coupled flows induced significant deviation from conduction dominated conditions, and the thickness of the fully solidified and mushy regions varied with vertical position. Furthermore,
as a result of variations in thermal and solutal conditions near the liquidus interface separating the mushy and melted zones, the shape of the liquidus interface was highly irregular. The streamlines in the plot reveal more clearly the thermally driven (counterclockwise) cell in the melt and the solutal driven (clockwise) cell in the mushy zone. Compared with the results in Example 1, the temperature and liquid composition fields have been distorted significantly by advection effects. The evolution of macrosegregation patterns was particularly studied. In Figure 4.19, there is a region where the liquidus penetrates into the mushy zone. Within this region, interdendritic fluid is channelled, and the subsequent effects are revealed by the mixture concentration plot of Figure 4.20.

4.2.3 Example 3

The effect of anisotropic permeability is studied in this example problem. In particular, the same geometry, physical parameters, initial and boundary conditions are considered as in Example 2. However, an anisotropic permeability model was used. Permeability in the mushy zone depended on whether the flow is parallel, perpendicular, or oblique to the growth direction of primary dendrites, which was assumed to be aligned with the local temperature gradient. In the present study, the ratio of the principle permeabilities \( R \) (parallel to perpendicular to temperature gradient) was taken to be 4, which means the permeability to flow in the vertical direction is relatively small. It is observed that the flow of water-rich interdendritic fluid to the top of the cavity is impeded, thereby retarding development of a double diffusive layer along the top of the liquid region. The significant influence of anisotropy on solidification suggests that the effect should be considered to achieve accurate simulation results.
4.3 Pure aluminum solidification

The versatility of the current program is also tested with an isothermal solidification problem. The test example is obtained from [37]. The problem definition is repeated here for completeness. A square mold with aspect ratio of 1.0 is considered. It is initially occupied by liquid Aluminum which is initially superheated by an amount of $\Delta T_o = 200^\circ C$. The ambient temperature was taken to be $25^\circ C$. A mixed temperature/flux boundary condition was applied to the left wall and all other walls were insulated. The dimensionless variables governing the problem are summarized in Table 4.3. A $40 \times 25$ finite element mesh was used for the computational region, consisting of 1000 nodes and 1066 elements. A dimensionless time step of $\Delta \tau = 0.0002$ is used for the whole simulation process. The results are illustrated in Figures 4.23.

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Prandtl number Pr</td>
<td>0.0149</td>
</tr>
<tr>
<td>Darcy number Da</td>
<td>$10^{-5}$</td>
</tr>
<tr>
<td>Rayleigh number Ra</td>
<td>$10^5$</td>
</tr>
<tr>
<td>Heat conductivity ratio $R_k$</td>
<td>1.0</td>
</tr>
<tr>
<td>Specific heat ratio $R_C$</td>
<td>1.0</td>
</tr>
<tr>
<td>Biot number Bi</td>
<td>3.3</td>
</tr>
</tbody>
</table>

Table 4.3: Dimensionless variables governing the solidification of pure Aluminum

Comparing the computed velocity and temperature fields with the results of [37], we can see that the position of the interface agrees very well with that obtained in [37], so does the flow pattern in the melt. As the solidification progresses, a counterclockwise circulation is established in the liquid region with the fluid ascending along
the right wall and cold fluid descending along the interface. From time $\tau = 0.35$, there is a tendency to form secondary cells near the upper end of the right wall. This cell evolved into a clockwise circulation at the top of the main flow. The good agreement shows that although the volume averaging method is developed for simulation of solidification processes with diffused mushy regions, it is still capable of simulating isothermal phase change problem with sharp interfaces. The change of liquid volume fraction from 0 to 1 here only spans one to two nodes.
Figure 4.7: Isothermal and Isoconcentration patterns of double diffusive convection in uniform porosity media
Figure 4.8: Geometry and boundary conditions of the natural convection problem with variable porosity
Figure 4.9: Isothermal pattern of natural convection in uniform porosity media
Figure 4.10: Geometry and boundary conditions of the solidification problem.

Figure 4.11: Computational grid for the solidification problem.
Figure 4.12: Solidification without convection at $\tau = 0.009$: (a) isothermals and (b) liquid isoconcentration lines.

Figure 4.13: Solidification without convection at $\tau = 0.018$: (a) isothermals and (b) liquid isoconcentration lines.
Figure 4.14: Solidification without convection at $\tau = 0.036$: (a) isothermals and (b) liquid isoconcentration lines.

Figure 4.15: Solidification without convection at $\tau = 0.07$: (a) isothermals and (b) liquid isoconcentration lines.
Figure 4.16: Solidification behavior at \( \tau = 0.009 \): (a) velocity vectors, (b) streamlines, (c) isothermals, (d) liquid isocomposition lines
Figure 4.17: Solidification behavior at $\tau = 0.036$: (a) velocity vectors, (b) streamlines, (c) isothermals, (d) liquid isocomposition lines
Figure 4.18: Solidification behavior at $\tau = 0.07$: (a) velocity vectors, (b) streamlines, (c) isothermals, (d) liquid isocomposition lines
Figure 4.19: Solidification behavior at $\tau = 0.14$: (a) velocity vectors, (b) streamlines, (c) isothermals, (d) liquid isocomposition lines
Figure 4.20: Macrosegregation pattern at $\tau = 0.14$
Figure 4.21: Solidification behavior with anisotropic permeability model at $\tau = 0.018$: (a) velocity vectors, (b) streamlines, (c) isothermals, (d) liquid isocomposition lines
Figure 4.22: Solidification behavior with anisotropic permeability model at \( \tau = 0.036 \): (a) velocity vectors, (b) streamlines, (c) isothermals, (d) liquid isocomposition lines.
Figure 4.23: Solidification of pure aluminum: dimensionless velocity and temperature fields for the case $Ra = 10^5$
Chapter 5

Suggestions for future studies

In this final chapter, we identify some open issues of interest for further research in the area of mathematical modelling of directional binary alloy solidification processes. Such important computational issues that need to be addressed include the following:

- Extension and improvement of the model to address the simulation of solidification systems with Rayleigh and Lewis numbers than those examined here. Using higher values of Rayleigh and Lewis numbers implies stronger melt convection with a very thin solutal boundary layer. This class of problems requires significant refinement in spatial and temporal discretizations to resolve the various features of the strong fluid flow. Increased memory requirements, storage and CPU costs make the flow problem in the strong convection regime highly challenging.

- Currently, the solution of the fluid flow subproblem is computationally very expensive. This high computational cost restricts the mesh size to which is unable to capture the details of the interfacial morphologies. Parallelization
of the program need to be considered to extend the methodology to address
the simulation of complicated three-dimensional solidification systems.

In the present study, only thermal-solutal convection was considered in analyzing
the macrosegregation pattern. As an extension of the application of volume averag-
ing technique, several other physical mechanisms which will affect macrosegregation
can be readily incorporated in the developed mathematical model. These physical
mechanisms include the following:

1. Thermo/diffusocapillary effect: Experimental and numerical results have shown
that surface tension caused by temperature and/or concentration gradients
along the free surface is another important driving force of fluid flow at a re-
duced gravity level. It is of significant interest to incorporate thermo/diffusocapillary
effect into the current model to study the relative importance of surface ten-
sion driven and buoyancy driven convection on the formation of segregation
patterns.

2. Shrinkage: In the present work, the density of the alloy was assumed to be
constant in both the liquid and solid except in the buoyancy terms. However,
it is well known that shrinkage is a source of interdendritic liquid motion in
alloy solidification [3, 4, 5]. An investigation of the conditions under which
shrinkage-induced flow significantly alters macrosegregation in a casting should
also be considered in the future.

3. Equiaxed solidification: The current model is developed for columnar den-
dritic solidification system where the solid is stationary. In equiaxed dendritic
solidification, the solid is moving with the fluid flow and will greatly change
the macrosegregation pattern in a favorable way. Additional supplementary
relation between the velocity of solid and the mixture velocity is needed in the deriva-
tion of such a model.

Both our study and other numerical and experimental results have shown that
buoyancy-induced fluid flow is primarily responsible for most forms of macrose-
gregation and that it affects the form and scale of the obtained solidification mi-
crostructures. Although convection is not directly controllable, there are some
means of eliminating buoyancy effects. In addition to mathematical models for
analysis of solidification processes, there is also significant need to develop experi-
mental techniques and computational algorithms that can allow the control of the
various physical mechanisms that are present in solidification. Such mechanisms are
the following:

1. Reduced gravity: Solidification in reduced or gravity-free environment is an ef-
fective method to control cast microstructures through suppression of buoyancy-
driven fluid flow. However, this approach is usually very costly or often im-
practical for many solidification systems and needs to be combined with other
control means (for example, optimally designed magnetic fields as well as ther-
mal boundary fluxes) to be economically feasible.

2. Electromagnetic field: The use of electromagnetic fields in materials processing
has been considered by many researchers [38]. Although effects of both steady
(dc) and time harmonic (ac) magnetic fields on solidification structure have
long been recognized [39, 40], the complex flow field and its interaction with
a dendritic mushy zone are not understood well enough.

3. Rotation: Experimental and numerical results have shown that centrifugal
forces induced due to rotation of the crucible/mold lead to enhanced crystal
properties through reduction in macrosegregation. Furthermore, recent studies [41, 42] have also demonstrated that intermittent rotation leads to suppression of the formation of channels, and therefore freckles, throughout most of a unidirectionally solidified casting.

4. Vibration: Recent experimental studies [43] have shown that strong vibration can be used effectively in suppressing channel and freckle formation in several aqueous solidification systems. Vibration can induce fine-scale mixing, thereby altering the effective diffusivities of species and heat and hence the effective Lewis number, which is an important parameter in double-diffusive convection [44].
Bibliography


