NUMERICAL MODELING OF AN ALLOY DROPLET DEPOSITION
WITH NON-EQUILIBRIUM SOLIDIFICATION

by

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Abstract

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Droplet deposition is a process of extensive relevance to the microfabrication industry. Various bonding and film deposition methods utilize single or multiple droplet impingements on a substrate with subsequent splat formation through simultaneous spreading and solidification. Splat morphology and solidification characteristics play vital roles in determining the final outcome. Experimental methods have limited reach in studying such phenomena owing to the extremely small time and length scales involved. Fundamental understanding of the governing principles of fluid flow, heat transfer and phase change provide effective means of studying such processes through computational techniques. The present study aims at numerically modeling and analyzing the phenomenon of splat formation and phase change in an alloy droplet deposition process. Phase change in alloys occurs non-isothermally and its formulation poses mathematical challenges. A highly non-linear flow field in conjunction with multiple interfaces and convection-diffusion governed phase transition are some of the highlighting features involved in the numerical formulation. Moreover, the non-equilibrium solidification behavior in eutectic systems is of prime concern. The peculiar phenomenon
requires special treatments in terms of modeling solid phase species diffusion, liquid phase enrichment during solute partitioning and isothermal eutectic transformation.

The flow field is solved using a two-step projection algorithm coupled with enhanced interface modeling schemes. The free surface tracking and reconstruction is achieved through two approaches: VOF-PLIC and CLSVOF to achieve optimum interface accuracy with minimal computational resources. The energy equation is written in terms of enthalpy with an additional source term to account for the phase change. The solidification phenomenon is modeled using a coupled temperature-solute scheme that reflects the microscopic effects arising due to dendritic growth taking place in rapidly solidifying domains. Solid phase diffusion theories proposed in the literature are incorporated in the solute conservation equation through a back diffusion parameter till the eutectic composition; beyond which a special treatment is proposed. A simplified homogeneous mushy region model has also been outline. Both models are employed to reproduce analytical results under limiting conditions and also experimentally verified.

The primary objective of the present work is to examine the splat morphology, solidification behavior and microstructural characteristics under varying operational parameters. A simplified homogeneous mushy region model is first applied to study the role of convection in an SS304 droplet deposition with substrate remelting. The results are compared with experimental findings reported in the literature and a good agreement is observed. Furthermore, a hypoeutectic Sn-Pb alloy droplet deposition is studied using a comprehensive coupled temperature solute model that accounts for the non-equilibrium solidification occurring in eutectic type of alloys. Particular focus is laid on the limitations of a homogeneous mushy region assumption, role of species composition in governing solidification, estimation of the microstructural properties and eutectic formation.
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Chapter 1

Introduction

Micro-droplet deposition process is continually being employed in small scale rapid manufacturing techniques. From micro-scale fabrication of solder bumps to larger 3D metal printing technologies, this process finds vital application in a range of modern day fabrication processes. Recent developments in this field are primarily focused on 3D printed parts through the use of metals and alloys. The major challenges in these developments are establishing stability of droplet generation, dealing with environmental effects on high temperature metal droplets and finding effective means for subsequent droplet fusions. On the other hand, well established processes such as solder bump formation on wafers used in electronic packages are also rigorously studied with varying objectives. Improved rigidity, material strength, optimum bump morphology and the search for a lead free material substitute are some of the many research interests requiring a robust understanding of droplet physics and its solidification characteristics. The droplet impacts are usually with a very small velocity and the resulting solidified part is called a bump. Applications of droplet deposition are not only limited to solder or metal bump formations. High velocity micro-droplet impingement is observed in processes like thin film deposition, thermal sputtering and spray cooling among others. The formation of multiple thin splats upon solidification results into a fine film created which requires minimum post fabrication treatment. A similar idea is used in Vapor Deposition Manufacturing (VDM), Plasma Deposition, Sputtering and other coating processes.

Major challenges in studying such processes experimentally are the extremely small time and length scales involved. Droplet morphology is greatly affected by the phase change process in addition to operating parameters such as impact velocity,
droplet size, initial droplet and substrate temperatures etc. Moreover, the estimation of material properties requires knowledge about the microstructural developments occurring within a droplet as it undergoes phase transformation. Experimental studies through High Speed Photography limit the investigations to superficial observations only. Microstructural observations carried out through SEM imaging techniques provide information about post solidification properties while the development phase remains unaccounted for. Since 1960’s scientists have been researching droplet physics, morphology and solidification characteristics with various methods and different objectives.

1.1 Background and Literature Review

The pioneering work in the modeling of phase change was the Classical Stefan problem [1]. It is believed that the particular work was motivated to study the formation of ice in polar regions. Mathematically, it is a differential equation given by Equation 1.1 describing the energy balance across a moving boundary undergoing solidification.

$$\rho L \frac{dy}{dt}\bigg|_{T=Tm} = J_l - J_s \quad (1.1)$$

where L is the latent heat of fusion, $J_l$ and $J_s$ represent the diffusion heat flux on the liquid and solid sides respectively and y is the location of the phase front. Solving the energy equation in the solid and liquid domains with above mentioned boundary condition yields the solution as

$$y = U\sqrt{at} \quad (1.2)$$

where U is a constant. The work was viewed as a ground breaking result in predicting the formation of ice in sea water and it was also experimentally verified. The result is till date used by scientists in studying various phenomenon ranging from microscopic dendritic
solidification in alloys to advancing phase front in isothermal solidification of metal castings.

The same expression was also adopted by Madejski [2] in 1960 to develop an analytical model to describe freezing affected droplet spread on a substrate. It was the first of a wide range of theoretical developments in studying droplet impingement and solidification. The basic assumption in his derivation was of a perfectly cylindrical splat immediately after impact and an assumed velocity profile representing the flattening of this disk shaped splat. This assumption eliminates the need to solve the flow governing equations. Moreover, the movement of the solidification front was inherited from Stefan’s solution given by Equation 1.2. Thus, the analytical description of $R(t)$ was such that it satisfied the energy equation given by:

$$\frac{d}{dt}(E_k + E_p + L_f) = 0$$

(1.3)

where $E_k =$ Kinetic Energy, $E_p =$ Potential Energy and $L_f =$ Viscous dissipation term. The detailed solution procedure can be found in [2]. An expression for spread factor $\xi = R/R_0$ was given for varying Reynolds number and Weber numbers.

![Splat-Quench Model](image-url)
A summary of these results and their evaluation is presented in [4]. It was found that the scheme gave highly accurate representation of the flattening of droplet for high Reynolds numbers but significant errors were observed otherwise. The main reason behind the failure to reproduce lower impact velocity cases was negligence of initial energy dissipation during impact (viscous dissipation and superheat) among others such as contact resistance, wetting effects and droplet recoil. Accuracy evaluation of this splat-quench model was performed by Aceves et al. [4] under different operating conditions. Spread factor and dimensionless splat thickness were predicted from the analytical model and compared with experimental findings. Numerous works have been published in the literature following Madejski’s splat-quench model with intentions of improving the flow dynamics (Markworth and Saunders [5], Trapaga et al. [6], Fukai et al. [7], Bennet and Poulakakos [8] and Rangel and Bian [3]) and solidification characteristics (Rangel and Bian [3], Attinger et al. [9], Amon et al. [10], Delplanque and Rangel [11], Kang et al. [12] and Collings et al. [13]).

Rangel and Bian [3] presented a substantially improved splat-quench model. One important aspect of their work was the solution of complete energy equation and the implicit tracking of the isothermal solid-liquid interface. This resulted in a robust analytical solution that could predict substrate remelting observed in rare situations. Moreover, this method also satisfied the potential energy conservation during the initial impact phase which was neglected in the previous model. Based on their findings, they also suggested a modification to the Madejski’s approach which claimed to improve the splat flattening accuracy. As Madejski’s assumed a cylindrical splay upon impact, \( \xi_0 = 0.5 \) (initial condition for spread factor) was arbitrarily assumed. If this condition is put in conjunction with the prescribed velocity profile, it can be proved that the energy conservation is violated during impact. Rangel and Bian [3] proposed using \( \xi_0 = 0.74 \) based on their full
energy equation solution. Aceves et al. [4] employed this in their accuracy evaluation and found approximately 30 % improvement in predicting the maximum spread factor. Delplanque and Rangel [14] later conducted a parametric study with a range of Webber and Reynolds number and compared their results with Madejski’s model. Significant deviations were observed for small values of \( \text{Re/We} \). It was concluded that for higher \( \text{We} \), surface tension dominates the initial impact and spread of the splat which had been overlooked in Madejski’s method.

Noteworthy contributions to the improvement and evaluation of these analytical models came from Trapaga et al. [6], Liu et al. [15, 16], Fukai et al. [7], Amon et al. [10] and others. However, one common assumption in all the above mentioned works was the assumption of an isothermal phase change. The solid-liquid interface was treated with a Stefan boundary condition either explicitly or implicitly. It implied that a multi-domain system of equations needed to be solved simultaneously while satisfying the interface condition. Numerical simulation with such assumption becomes very difficult as it necessitates the tracking and reconstruction of a moving boundary in addition to coupling multiple numerical solutions. Pasandideh-Fard et al. [17, 18] performed numerical analysis of the process with different objectives including the study of heat transfer and splat morphology. Liu [19] summarizes their findings. It can be interpreted from these works that modeling an isothermal phase change in conjunction with droplet impact and deformation requires extremely complex numerical techniques and highly restrictive theoretical assumptions. Moreover, isothermal solidification is physically possible only in pure metals. State-of-the-art fabrication technologies make use of sophisticated metal alloys which exhibit superior strength, environmental stability and anti-oxidation properties. Therefore, it is vital to be able to simulate, analyze and study the solidification characteristics of alloys undergoing non-isothermal phase change.
Researchers in the past have solved the conventional energy equation under varying domain properties to simulate accurate thermal profiles. Although, the problem of phase change had largely been treated as an interface condition rather than a continuum. The pioneering work of Brent et al. [20] made it possible to express the phase change phenomenon in a continuum domain. The energy equation was expressed in terms of total enthalpy rather than just sensible heat. The total enthalpy is a sum of the latent heat content and the sensible heat. The modified form of the energy equation was called (and will now on be referred to as) the enthalpy formulation. Solution of the enthalpy formulation involves two unknowns: sensible heat and latent heat. Therefore, additional information is needed to express a relation between these two quantities. Assumption of a linear relationship between the two quantities was suggested. This situation typically occurs in solidification of alloys where distinct liquidus and solidus temperatures are determined from alloy composition and phase diagrams. The range of temperatures between which phase change occurs is called the Mushy Region. Application of this method is a part of the present work and will be elaborated in later sections. The method provides a numerically convenient technique to model phase change. The approach has now become a widely accepted means of analyzing phase change in almost any given domain. Commercial thermal and flow simulation packages also use this method to simulate phase change. Moreover, this method can be conveniently employed in applications involving alternating melting and solidification cycles such as PCMs in thermal energy storage and heat sinks.

In addition to these applications, the method can easily incorporate simultaneous effects of flow field on a solidifying domain. Shrinkage driven flow and convection driven phase change are examples where it is important to couple the flow field with the governing equation for phase change i.e. enthalpy formulation. Droplet deposition is one
of the most challenging situations where this method proves highly efficient. Two of the proposed means of coupling flow field with enthalpy formulation are the pseudo porosity method [20] and viscosity manipulation [21]. Detailed description of the pseudo porosity model is given in later sections.

The idea was implemented in simulating droplet solidification by Zarzalejo et al. [22] and Schmaltz [23]. This technique was used in their studies relating convection effects in droplet solidification and microstructural examinations. Substrate remelting was also easily achieved through this method. However, the full potential of this technique was not exposed in their work because convection was indirectly treated with an artificially augmented conductivity to mimic the effects of flow field. The fundamental problem behavior was still a diffusion-only type. Tong and Holt [24] used the enthalpy-porosity approach in studying a single copper droplet deposition on a substrate by numerically solving the complete energy equation inclusive of convection terms. The isothermal solidification front was interpreted as an infinitesimally small mushy region. A dimensionless analysis incorporating the effects of superheat, impact velocity and substrate preheating was reported. Later, substrate remelting, convection effects and microstructural properties of an SS splat were examined using a similar method by Ramanuj et al. [25]. Enthalpy-porosity technique proves to be an efficient numerical method to simulate any phase change process. However, the assumption of a linear relationship poses limitations in accuracy when predicting certain thermo-physical properties of the solidified splat. Examination of these limitations is a part of the present work and will be elaborated in subsequent chapters.

A summary of the literature review outlined here is presented in Table 1-1.
<table>
<thead>
<tr>
<th>Model</th>
<th>Flow Field Solution</th>
<th>Free Surface Tracking</th>
<th>Phase Change</th>
<th>Concentration Consideration</th>
<th>Contribution</th>
</tr>
</thead>
<tbody>
<tr>
<td>Madejski [2]</td>
<td>N/A (Assumed)</td>
<td>N/A (Cylindrical Splat)</td>
<td>Isothermal, Stefan</td>
<td>N/A</td>
<td>Solution of energy equation for droplet spreading</td>
</tr>
<tr>
<td>Markworth and Saunders [5]</td>
<td>N/A (Assumed)</td>
<td>N/A (Cylindrical Splat)</td>
<td>Isothermal, Stefan</td>
<td>N/A</td>
<td>Modified Madejski with flow field to satisfy zero shear at the free surface</td>
</tr>
<tr>
<td>Delplanque and Rangel [11]</td>
<td>N/A (Assumed)</td>
<td>N/A (Cylindrical Splat)</td>
<td>Isothermal, Stefan</td>
<td>N/A</td>
<td>Modified Madejski to include potential energy change after impact</td>
</tr>
<tr>
<td>Aceves et al. [4]</td>
<td>N/A (Assumed)</td>
<td>N/A (Cylindrical Splat)</td>
<td>Isothermal, Stefan</td>
<td>N/A</td>
<td>Modified Madejski to account for viscous dissipation during initial spreading</td>
</tr>
<tr>
<td>Collings et al. [13]</td>
<td>N/A (Assumed)</td>
<td>N/A (Cylindrical Splat)</td>
<td>Isothermal, Stefan</td>
<td>N/A</td>
<td>Modified Madejski to account for surface tension in the energy conservation</td>
</tr>
<tr>
<td>Rangel and Bian [3]</td>
<td>N/A (Assumed)</td>
<td>N/A (Cylindrical Splat)</td>
<td>Isothermal</td>
<td>N/A</td>
<td>Implicit solid-liquid interface tracking with remelting</td>
</tr>
<tr>
<td>Zarzalejo et al. [22]</td>
<td>N/A (Flow Field Absent)</td>
<td>N/A (Prescribed, Fixed)</td>
<td>Non-isothermal</td>
<td>N/A</td>
<td>Augmented diffusion limit model with substrate remelting</td>
</tr>
<tr>
<td>Liu et al. [15]</td>
<td>Two-Step Projection</td>
<td>VOF (SLIC)</td>
<td>Isothermal, Stefan</td>
<td>N/A</td>
<td>Numerical model with full NS equation and solidification taken from Madejski</td>
</tr>
<tr>
<td>Pasandideh-Fard [18]</td>
<td>SOLA</td>
<td>VOF (Donor-Acceptor)</td>
<td>Isothermal, Approximate</td>
<td>N/A</td>
<td>Numerical model with improved solidification</td>
</tr>
<tr>
<td>Tong and Holt [24]</td>
<td>Two-Step Projection</td>
<td>VOF (SLIC)</td>
<td>Non-isothermal</td>
<td>N/A</td>
<td>Numerical model with convection-diffusion governed energy equation</td>
</tr>
<tr>
<td>Attinger and Poulidakos [26]</td>
<td>FEM</td>
<td>Multi-domain approach</td>
<td>Isothermal</td>
<td>N/A</td>
<td>Numerical Finite Element Model to incorporate multiple droplet impacts</td>
</tr>
</tbody>
</table>
Evidently, analytical solutions came with the primary limitation of decoupled flow field and solidification process. The numerical models were able to couple the two phenomena and provide improved accuracies. However, all those studies do mention the negligence of species composition in governing the phase change process. Temperature variations in non-isothermal phase change are not solely governed by the energy conservation. Solidification is a complex phenomenon involving a number of microscopic governing factors indirectly affecting the thermal behavior. Phase transformation has remained a popular research area among material scientists since decades. Theoretical developments in this field focus on identifying the thermo-solutal coupled relationships so as to have a mathematical representation to accurately predict solidification and microstructural properties. Phase diagrams of binary alloys graphically represent the equilibrium conditions between thermodynamic quantities and phase specific properties. These charts are based on the minimum stable energy state required for specific atomic arrangements (phases). Phase diagrams may be the simplest forms of establishing the thermo-solutal relationship. However, quasi-equilibrium states do exist based on cooling conditions governed by temperature gradients and cooling rates. A plethora of classical solutions [27, 28, 29, 30, 31, 32] have been proposed in literature to express thermo-solutal coupling under varying circumstances.

Stability of solid-liquid interface is characterized primarily by temperature gradient and interface velocity. [33] gives a map of different interface conditions based on the above mentioned criteria. Note that pure metals always have a stable planar interface which is the reason for isothermal solidification. Binary alloys on the other hand may exhibit planar (rare) and dendritic/columnar growth which is an indirect outcome of the thermo-solutal coupling. Cooling rate typically in excess of $10^3$ K/s fall under the category of rapid solidification and show dendritic microstructures. While the equilibrium
solidification becomes violated, analysis of rapid solidification is fairly complicated due to factors such as segregation, dendritic arm coarsening and finite solid state species diffusion.

A whole new challenge is posed for numerical research in modeling the phase change process owing to the extremely small length scale on which these theories are based. Thermo-solutal relationships are described on a scale as small as $10^{-6}$ m (inter-dendritic spacing) whereas most phase change processes occur at a scale at least $10^{3}$ times larger than that. Moreover, classical theories are based on certain fundamental assumptions limiting the applications to hypothetical situations. Practical applications require simplification of the real domain (e.g. Madejski’s model) to a great extent and thereby compromising the accuracy to mimic actual processes. Attinger et al. [9] describe the scaling issues in this respect in their summary of droplet physics. The need to have a generalized formulation for phase change has also been emphasized. Numerical challenges in modeling this phenomenon have motivated scientists to research means of extending and improving the reach of the thermo-solutal coupling theories. Eutectic phase diagrams are the simplest models representing equilibrium solidification of binary alloys. The classical lever rule [31] is widely used to relate temperature with solute composition for an alloy undergoing phase transition. Processes such as a droplet deposition where extremely high cooling rates are involved do not follow the equilibrium condition. Moreover, the microstructural observations of parts produced through rapid solidification exhibit eutectic lamellar structure. The formation of such microstructure was explained through the rapid solidification model given by Scheil-Gulliver theory [31]. Improvements to these pioneering works are explained in later sections. Through these developments, phase transition studies through analytical methods have been made possible. Although, these methods are only applicable under a very restrictive conditions.
Solute transport through advection and the role of convection in the energy equation play a vital role in processes such as solder jetting, thermal sputtering and micro-droplet deposition.

Ni and Beckerman [34] proposed the concept of Representative Elementary Volume (REV) to facilitate the application of established theories on a macro-scale simulation. Voller et al. [35] later used the method to incorporate convection effects through the use of his enthalpy-porosity technique. Based on his model, a similarity solution for one dimensional shrinkage driven flow [30] was also developed. Important contributions of these works have been adopted in the present study and will be discussed in subsequent sections. Timchenko et al. [36] developed a coupled temperature-solute model for computational studies of the famous MEPHISTO experiment. The model was used to examine the growth properties and solidification behavior under micro-gravity environment.

Numerical studies on binary alloy solidification have been made possible through Voller’s [37] approximate back diffusion treatment. The classical results only work under specific assumptions of growth profile. It is not practical to utilize the results selectively under varying circumstances. Dendritic solidification is a complex microscopic phenomenon observed in alloy solidification. The coupled temperature-solute relationship makes it challenging to model the phenomenon numerically. Moreover, diffusion in solid state, although minimal, governs the development of the dendritic surface to a great extent. Treatment of this solid state diffusion is a characteristic of the classical solutions. The surface equilibrium condition governing the advancement of the dendritic surface is clearly dependent on the treatment of this quantity. The approximation generalizes the phenomenon through a simple formulation which can easily be incorporated in the
numerical scheme. It has to be noted that exact modeling of solid state diffusion is not possible through a macroscopic model.

As mentioned earlier, eutectic type of alloys exhibit a peculiar solidification behavior. All the theories mentioned above are only valid within the mushy region. Figure 1-2 shows a simple phase diagram of a binary alloy.

![Eutectic equilibrium diagram with solid solution](https://www.subtech.com)

**Figure 1-2 Example of a Binary Eutectic Phase Diagram**

Assuming C to be the initial concentration at the initial point M, T₁ is the liquidus temperature marking the onset of phase change. Note that temperature and concentration are not independent quantities below T₁. A decrease in concentration from T₁ to T leads to enrichment of the liquid mixture. The process continues till C_y = C_{eut} known as the eutectic point. This is a unique point in the phase diagram marked by (C_{eut}, T_{eut}). Further cooling leads to the formation of a eutectic phase which solidifies isothermally. Therefore, the coupled treatment encounters a discontinuity at the eutectic composition. All the classical theories [27, 28, 29, 31, 32] are only valid till this point. Voller’s method [37, 38] suggested extrapolation of the liquidus curve below eutectic and
assuming no back diffusion. Although mathematically possible, the method violates the physics of eutectic formation. It has to be noted that most of the times detection of eutectic microstructure is the primary objective and the existing models do accomplish that. However, solidification analysis, morphological studies and microstructural estimations require a complete simulation. This has remained one of the major limitations of all the existing theories.

1.2 Motivation and Objective

Numerical simulations often provide a detailed view of the overall process which experimental analysis may not be able to. Droplet deposition manufacturing is one such process where experimental observations are very difficult owing to the extremely small time and length scales involved. Visualization techniques for such processes are very rare and expensive. Peripheral observation is not sufficient for detailed study and optimization. Flow profiles, interface advancement and thermodynamic behavior are the governing phenomenon in a droplet deposition which are not possible to monitor through experimental methods. Moreover, the microscopic phenomena such as back diffusion, dendritic growth, solute separation are only detected post solidification and through imaging techniques which require alteration of the formed part. Continuous development of such quantities is rather impossible. Knowledge of governing principles i.e. mass, momentum and energy conservation in conjunction with state-of-the-art computational facilities enables one to simulate the entire process and also obtain visual as well as quantitative information throughout the entire process. Such analysis can be used in identifying the crucial governing elements and operational parameters to optimize the process for the desired outcome with minimal resources.
Numerical modeling of droplet impingement and solidification is highly challenging. As mentioned earlier, droplet impact needs to be accurately modeled to obtain a precise description of the subsequent events. The basic challenge involved here is to track and reconstruct multiple deforming interfaces. In addition, the governing equations for different media need to be solved through a continuum approach. The classical governing equations need to be modified to incorporate the phase change. Moreover, solidification is governed by the coupled energy and solute conservation equations. The eutectic behavior makes it even more challenging. Accurate simulation of eutectic solidification requires special treatment.

The primary objective is to study the phenomenon of phase transition observed in a droplet undergoing simultaneous deformation and solidification. The droplet deposition can be categorized into two aspects taking place and affecting each other simultaneously: Impact/deformation and solidification. The numerical model described here is intended to capture the simultaneous spread and phase change of the droplet. The resulting splat formation is examined through morphological observation as well as detailed quantitative analysis of solidification characteristics. The role of droplet spread and dimensionless operating parameters on cooling, solidification and microstructural properties is also studied with particular focus on eutectic formation. A comparison with a simplified phase change model is presented to emphasize the role of coupled temperature-solute relationship in governing solidification. Validation of various aspects of the numerical model is also presented. Effects of alloy composition and material properties are analyzed to examine the capabilities and limitations of both models. The mathematical formulation and numerical solution algorithm are briefly described below followed by model validation and results and discussion. The scope of further improvements and applications of the present work is also given.
Chapter 2
Mathematical Formulation

As mentioned in the previous section, the solution for flow field, interface tracking and reconstruction, modeling of phase change and solution of the energy equation are the primary aspects of the numerical algorithm. In general, the continuity equation, Navier-Stokes equation and the energy equation need to be solved simultaneously. However, the presence of multiple interfaces and phase transformation make the process more challenging. A finite volume approach on a fixed grid is used here for the discretization of the governing equations. The formulation of the governing equations on a continuum domain is discussed here along with various aspects of solidification. The variations of the thermo-fluid properties (excluding specific heat) with temperature throughout the scope of the present study have been neglected to emphasize the effects of solidification models discussed later.

2.1 Flow Governing Equations

The classical continuity and Navier-Stokes equations govern the flow field. However, due to the presence of multiple phases within the domain the governing equations need to be modified as follows:

\[ \nabla \cdot \vec{V} = 0 \]  \hspace{1cm} \text{(2.1)}

\[ \frac{\partial \vec{V}}{\partial t} + \nabla \cdot (\vec{V} \vec{V}) = -\frac{1}{\rho} \nabla P + \frac{1}{\rho} \vec{\tau} + \vec{g} + \frac{1}{\rho} \vec{F}_b + S_v \]  \hspace{1cm} \text{(2.2)}

Note that the density variation terms from both equations have been dropped because the flow is considered incompressible. However, shrinkage effects can be accounted by the Boussinesque approximation in the body force terms.
\( \theta \) is a quantity defined as liquid fraction which denotes the volume fraction of liquid within a given control volume. The basic assumption in the formulation is that solid particles are treated as a continuous stationary medium. The region where solid as well as liquid media are present is defined as mushy region. Quantification of the liquid fraction and modeling of mushy region is dependent on the phase change process and will be discussed later. For the purpose of determining the flow field, solid portion is considered as a blockage within a flowing fluid medium. Thus, mushy region can also be modeled as a porous medium where porosity is determined from the solidification model. The term \( S_v \) in Equation 2.2 represents pressure impedance experienced through blockage. The classical Darcy's theory for flow in a porous medium states that the deceleration due to blockage can be written as:

\[
S_v = -AV \quad \text{and} \quad A = -C \frac{(1 - \theta)^2}{(\theta^3 + \epsilon)}
\]  

which is also called Carmen-Kozeny equation [39]. \( \epsilon \) is an arbitrary small number to avoid division by zero. ‘\( C \)’ is called the mushy constant. The works of [40] discuss the means of determining this value. It is reported that in case of dendritic solidification the mushy constant is inversely proportional to the dendritic length scale, empirically relate it as:

\[
C = \frac{180}{\lambda^3}
\]  

where \( \lambda \) is the interdendritic spacing which is of the order of \( 10^{-6} \) to \( 10^{-9} \) m. Typically, a mushy constant in the range \( 10^{9} \) to \( 10^{12} \) give satisfactory results in most applications. It is worth mentioning that some researches [21, 41] represent the blockage effects using a different approach where fluid viscosity is made a function of temperature such that it assumes a very high value in solid portions forcing the velocity to go to zero. However, the phase change model proposed here is compatible with the mushy region assumption.
and makes the formulation simpler. Moreover, viscosity manipulation makes the modeling of free surface difficult and is not a practical option in interfacial flows.

Assuming metals and alloys as Newtonian fluids, the viscous stress in Equation 2.2 is given by

\[ \tau = 2\mu S \quad \text{and} \quad S = \frac{1}{2} \left[ \nabla V + (\nabla V)^T \right] \]

where \( S \) is the rate-of-strain tensor. Taking the fluid-gas interface as a shear-free surface, the scalar boundary condition can be written as

\[ \Delta P_s = \sigma K \]

where \( K \) denotes the local curvature and \( \sigma \) is the surface tension. Note that \( \Delta P_s \) is the jump in fluid pressure across the interface and poses a discontinuity. A continuum surface force method [42] is employed to translate the surface boundary condition into a body force distributed across a finite thickness near the free surface. Numerical calculation of this term will be described in the subsequent section.

2.2 Interface Tracking Schemes

A wide range of applications in micro-fluidics, droplet phenomenon, open channel flows and surface wave propagations demand for a robust interface simulation technique. Experimental research in these fields has been performed through imaging methods such as PIV and LDV where visual, qualitative information is acquired and translated into quantified data through interpolation schemes. In recent years, extensive research is being conducted to simulate fluid interfaces through computational methods. Mathematical formulation of interfaces is broadly classified into Eulerian and Lagrangian methods. A brief review of these approaches, their advantages and limitations has been discussed here along with popular techniques proposed in the literature.
Lagrangian methods track the interface explicitly through solving the basic equation of motion given by

\[ \frac{d\vec{s}}{dt} = \vec{V} \]  

(2.7)

where \( \vec{s} \) is the position vector and \( \vec{V} \) is the velocity vector of the interface point. The use of this equation is explained through the following techniques.

**Moving Grid Method**

These methods involve a multi-domain approach for the solution of governing equations. Different sets of equations are individually solved in each domain and the interface poses a boundary condition to link the solutions. Explicit definition of the interface is obtained through Equation 2.7. Finite element analysis in fluids use this approach as it becomes easier to define the free surface as a set of discrete elements governed by the above equation. [43, 44] describe detailed application of this method. Generally, problems with regular topology undergoing minor deformations such as open channel flows, surface wave propagations and certain fluid-structure interacts can be accurately simulated. Drastic changes may lead to violation of element connectivity rules.

**Front Tracking Method**

The front tracking method originates from the famous Marker-and-Cell (MAC) method and subsequent modifications made by Daly [45] to implement it numerically. Discrete Lagrangian markers are assumed at the interface moving through a Eulerian grid. Motion of these markers is governed by Equation 2.7. Higher order interpolation polynomials are used to construct a curve representing the interface at each instant. While the interface is smooth and can handle drastic topological deformations, its definition is artificial rather than physical. Accuracy of the interface simulation highly depends on the interpolation scheme used. Poor mass conservation is another limitation.
Particle-Based Method

This is an example of grid-less simulation method which was originally developed to analyze astrophysical problems [46]. The basic ideal behind this method is to represent a continuum fluid as a collection of discrete particles carrying mass, momentum and energy. Multi-fluid problems are easily handled by describing heavier fluid as particle and lighter fluid as void. Thus, interface definition is automatically created. Although, fluid properties are weighted average of neighboring particles. Mass conservation is exact while the overall accuracy is limited to a great extent. Particle-in-cell (PIC) or smoothed-particle-hydrodynamics (SPH) partially or completely eliminate the meshing of the domain, thereby mitigating grid dependency of the solution.

Eulerain methods treat the interface through a continuum approach. An appropriate field function ‘C’ defines the location of the interface and is advanced in time under the effect of the flow field by solving the following advection equation.

\[
\frac{DC}{Dt} = \frac{\partial C}{\partial t} + \vec{V} \cdot \nabla C = 0
\]  

(2.8)

The interface is implicitly captured and reconstructed using the updated field function values. Interface boundary condition is completely eliminated in these methods. Moreover, such an approach facilitates implicit numerical evaluation of certain surface phenomenon as demonstrated in later sections.

Continuum Advection Method

This method involves the solution of Equation 2.8 through finite difference schemes. Such schemes are based on the assumption that the field function is continuous across the interface and consequently result into numerical diffusion problems. Although, cases where the fluid domain is large compared to the interface can be simulated accurately when the length scale of interface diffusion is negligible compared to overall problem dimensions [47]. Small scale fluid dynamic problems such
as droplet deformations spray formation and micro-fluidics may result into unrealistic interfaces through the use of this technique.

**Volume Tracking Method**

Volume tracking is probably the most accepted and universal Eulerian approach to simulate interface deformations. The interface is captured through a volume of fluid function representing the fractional fluid volume in a particular control volume. Post advection interface is reconstructed from the VOF values geometrically. Various developments [48, 49, 50, 51] are proposed in the literature to improve the interface accuracy using this method. Implicit mass conservation property is a salient feature of this method. However, orientation of the interface may not be highly accurate as the spatial derivatives of VOF function are not continuous near the interface.

**Level Set Method**

Osher and Sethian [52] proposed the level set method for capturing fluid interfaces but in recent years, its application has also been extended to efficient grid generation. The interface is defined by a signed distance function whose magnitude indicates the shortest distance of the interface from the cell center. The value is negative inside and positive outside the fluid. The level set function, unlike VOF, is a continuous function throughout the domain which means that spatial derivatives of the field function are highly accurate. However, once advected, it fails to retain the definition as being a distance function. Re-initialization of the level set function is necessary at each time step and in the process, mass conservation is not closely satisfied.

The present numerical model is based on a finite differencing scheme for flow and energy governing equations. As outlined above, Eulerian methods are compatible with this specific type of discretization. Moreover, the extremely small length scales involved in the problem make volume tracking as an ideal interface simulation technique.
Formulation of the scheme is described here while numerical solution method is coupled with the flow solver and will be presented in the next section. An enhanced scheme called the Coupled Level Set and Volume of Fluid Method (CLSVOF) proposed by [53] is also briefly reviewed.

2.2.3 Volume of Fluid Method (VOF)

The field function in Equation 2.8 is replaced with the volume of fluid (VOF) function, \( F \), defined as:

\[
F = \frac{\text{Volume occupied by fluid}}{\text{Total volume of the computational cell}}
\]  

(2.9)

Following information can be interpreted from this definition:

\[
F = \begin{cases} 
1 & \text{Completely Filled} \\
0 < F < 1 & \text{Partially Filled} \\
0 & \text{Void}
\end{cases}
\]  

(2.10)

Equation 2.8 with \( C = F \) is expressed in conservative form to ensure mass conservation.

\[
\frac{DF}{Dt} = \frac{\partial F}{\partial t} + \vec{V} \cdot \nabla F = 0
\]  

(2.11)

To better understand the implications of a VOF function, consider a hypothetical circular fluid element as shown in Figure 2-1. The domain is divided into square grids representing a control volume. The value prescribed in each cell is its corresponding VOF function. The image conforms to the implications mentioned earlier. Once the VOF distribution is acquired in the whole domain, it is advected using Equation 2.11. Numerical solution of the advection equation will be described later. Post advection, the interface is lost and needs to be reconstructed from the updated field function values.
A Piecewise Linear Interface Construction (PLIC) [54] scheme is adopted here. For example, the interface definition for the case shown in Figure 2-2 is given by:

\[ y = m \cdot s + y_{\text{int}} \quad (2.12) \]

where ‘m’ is the slope obtained from the normal vector, \( s \) is the x-coordinate (x for Cartesian and \( r \) for Cylindrical) and \( y_{\text{int}} \) is the intercept.
Note that Figure 2-2 is just one of the sixteen different possibilities in which the interface within a cell could be oriented. The numerical algorithm proposed by Rudman [49] outlines a cell rotation and indexing routines through which each of the orientations can be transformed such that the unit normal always lies in the first quadrant and Equation 2.12 gives the interface. ‘y’ and ‘s’ are the transformed coordinates corresponding to ‘z’ and ‘r’ in cylindrical systems. The normal vector is computed as

\[
\hat{n} = \frac{\nabla F}{|\nabla F|}
\]  

(2.13)

where \(|\nabla F| = 1\).

2.2.4 Coupled Level Set Volume of Fluid Method (CLSVOF)

The coupled level set method was proposed by Tong and Wang [53] with an intention of preserving the mass conservation property of the interface tracking scheme as well as obtaining an accurate interface orientation. This was done through coupling the VOF and LS methods as described in [55]. The LS method is introduced here followed by a summary of the overall scheme.

\[
\varphi = \begin{cases} 
< 0 & \text{Cell center is inside the fluid} \\
= 0 & \text{Cell center is at the interface} \\
> 0 & \text{Cell center is outside the fluid}
\end{cases}
\]  

(2.14)

An example of the LS distribution within a computational domain is given in Figure 2-3 and the following information can be interpreted from the same. Taking this as the field function in Equation 2.8 and advecting it results into a new set of LS values. Although, as indicated earlier, the updated field function values do not retain the same physical meaning i.e. distance function. Therefore, a re-distancing routine proposed in [55] is required.
Once the VOF and LS functions are advected, accurate interface location is tracked from the VOF function. Orientation of the interface is determined from the LS function as:

\[ \hat{n} = \nabla \varphi \]  

which is more accurate as compared to Equation 2.13. The slope in Equation 2.12 is replaced with the corresponding value.

### 2.3 Energy Equation

The conventional energy equation is given in terms of temperature as:

\[ \frac{\partial T}{\partial t} + \nabla \cdot (\bar{V} T) - \nabla \cdot (\alpha \nabla T) = 0 \]  

Phase change problems involve consideration of latent heat addition or dissipation from the system. Therefore, the above equation needs modifications to be applicable in the present work. Voller et al. [56] suggested using the total enthalpy in the energy equation.
rather than just sensible heat. The total enthalpy $H$ is given as the sum of sensible heat $(h)$ and latent heat $(\delta h)$. Equation 2.16 can now be modified as:

$$\frac{\partial H}{\partial t} + \nabla \cdot (\vec{v} H) - \nabla \cdot (\alpha \nabla h) = 0 \quad (2.17)$$

and re-grouped as:

$$\frac{\partial h}{\partial t} + \nabla \cdot (\vec{v} h) - \nabla \cdot (\alpha \nabla h) + \frac{\partial \delta h}{\partial t} + \nabla \cdot (\vec{v} \delta h) = 0 \quad (2.18)$$

Note that diffusion terms in Equation 2.18 remain unchanged as the diffusion flux is only governed by the temperature gradient. Convection flux, on the other hand, involves the total enthalpy.

Pure metals have a unique temperature, called fusion temperature, at which complete latent heat is released (solidification) or absorbed (melting). Such a behavior poses a discontinuity in Equation 2.18 at the fusion temperature. Therefore it becomes impossible to model isothermal phase change in materials through continuum approach. The method suggested in [24] is to approximate the phase change front as a thin mushy region of finite temperature range over which the latent heat is distributed. A qualitative enthalpy-temperature relation assuming constant specific heat is show in Figure 2-4. The infinite $dh/dT$ slope for an isothermal enthalpy jump is replaced with a finite slope (having a large value) confined within the temperature range $T_s$ to $T_l$.

The liquid fraction is defined as the fractional fluid volume in liquid state within a control volume. Mathematically, it can be expressed as the normalized latent heat given by:

$$\theta = \frac{\delta h}{L} \quad (2.19)$$

Computation of this quantity requires information about the phase change process and will be discussed later in the section.
Following interpretation can be drawn from the value of liquid fraction:

\[
\theta = \begin{cases} 
0 & \text{Matter is in solid phase} \\
0 < \theta < 1 & \text{Matter is in mushy region} \\
1 & \text{Matter is in liquid phase}
\end{cases}
\] (2.20)

Replacing \( \delta h \) with this definition in Equation 2.18 yields the following:

\[
\frac{\partial h}{\partial t} + \nabla \cdot (\vec{V} h) - \nabla \cdot (\alpha \nabla h) + S_h = 0
\] (2.21)

where

\[
S_h = L \left( \frac{\partial \theta}{\partial t} + \nabla \cdot (\vec{V} \theta) \right)
\] (2.22)

Note that the first three terms are exactly similar to the conventional energy equation while the remaining terms play the role of energy generation or dissipation in the form of latent heat during phase change. It can be understood that the traditional energy equation is regained in pure liquid or pure solid phases. The source terms become active only within the mushy range. Also note that this is the term appearing in the modified
continuity and Navier-Stokes equation representing porosity. If the solid particles are arbitrarily assigned zero velocity, the volume averaged velocity within a control volume can be given by:

\[
\vec{V} = \theta \vec{V}_l
\]  

(2.23)

where \(V_l\) is the fluid velocity determined from the flow governing Equation. Determination of the term \(\theta\) is of prime importance which will be discussed in later sections.

2.4 Non-Equilibrium Phase Transformation

Phase change phenomenon can occur in different ways depending on the type of material. As mentioned above, pure metals undergo isothermal phase change while alloys usually exhibit non-isothermal solidification. The scope of the present study being alloy solidification, the following discussion will pertain to non-isothermal solidification only. The work of Tong and Holt [24] on a metal droplet deposition describes how isothermal solidification can be approximated through an infinitesimally small mushy range. Alloys can be treated as solid solutions to understand the factors leading to a mushy region. In the example of a binary alloy, the two constituent species may have varying affinity or solubility limit for the other component. The free energy analyses of phase change in alloy reveal that the composition and thermodynamic state are dependent quantities under equilibrium condition.

Eutectic and Isomorphous are the two most commonly encountered binary alloy systems in material applications. The difference between the two systems is that maximum solubility limit for the primary solid phase, \(\alpha\) does not exist in isomorphous alloys and consequently, they show continuous liquidus and solidus curves. Eutectic reaction however, is a peculiar case where there is a finite solid state solubility limit in at
least one of the solid phases. Figure 2-5 show various configurations possible corresponding to the minimum free energy state of the system [57].

Figure 2-5 Equilibrium Free Energy Configurations

Variations from a eutectic reaction are the situations where a metastable state is formed (e.g. Al-Cu). Although being different from a standard eutectic system, the thermodynamics of phase change follow the same fundamentals for all eutectics. Therefore, energy equation and solute conservation are coupled governing principles in an alloy system undergoing phase transition. Moreover, rapid solidification as observed in droplet depositions deviates from the equilibrium assumption and needs special consideration.

As observed from the phase diagram, the liquid becomes enriched in solute as the solidification progresses. The ratio of solid to liquid concentration within the mushy region is called the solute partitioning coefficient given by:

\[
kp = \frac{C_s}{C_l} = \frac{f_l^{-1}(T_{equ})}{f_s^{-1}(T_{equ})} \tag{2.24}
\]

where \( T_{equ} \) is the equilibrium temperature and \( f_s \) and \( f_l \) are the functions defining the solidus and liquidus curves in terms of the solid and liquid concentrations \( C_s \) and \( C_l \).
respectively in the equilibrium phase diagram. The definition of these curves becomes simpler in eutectic systems as the non-linearity is negligible and a straight line assumption has been employed by various scientists to achieve satisfactory results [30, 37, 58]. With similar assumption,

\[ T_{\text{equ}} = T_{\text{fus}} + m_l C_l = T_{\text{fus}} + m_s C_s \]  

(2.25)

define the liquidus and solidus respectively with \( T_{\text{fus}} \) being the fusion temperature of pure substance. In that case, the partitioning coefficient can also be viewed as the ratio of the slopes of the liquidus to solidus lines in the phase diagram. Note that the linear assumption does not work in case of isomorphous alloys.

2.4.1 Homogeneous Mushy Region

The works of [20, 24, 25, 56] assume an approach called the homogeneous mushy region method where the energy equation is solved independently. The role of species composition in determining the thermodynamics of phase change is decoupled from the energy equation using certain assumption. The solidus and liquidus temperatures at a particular initial alloy composition can easily be obtained from the equilibrium phase diagram shown above. The mushy region is taken as a continuum with uniform composition. Such an assumption eliminates the need of formulating the solute conservation equation. Moreover, the latent heat is assumed to evolve uniformly between the two bounding temperatures for the mushy region obtained from the equilibrium phase diagrams. Mathematically, the assumption can be written as:

\[ \theta = \frac{T_m - T_s}{T_l - T_s} \]  

(2.26)

where \( T_m \) is the temperature within the mushy region. Equation 2.26 can be used in Equation 2.18 and the unknown \( \delta h \) can be eliminated. Moreover, if the material
properties are considered constant, the following equation can be derived where the specific heat is artificially augmented by a factor, $\frac{y+1}{y}$ within the mushy region.

$$\frac{\partial H}{\partial t} + \nabla \cdot (\bar{v} H) - \nabla \cdot (\alpha \gamma \frac{y}{y+1} \nabla H) = 0 \quad (2.27)$$

$$\gamma = \frac{c_p(T_l - T_s)}{L} \quad (2.28)$$

represents the ratio of sensible to latent heat released during phase change.

2.4.2 Non-Equilibrium Solidification

Equilibrium state in solidification is defined as the state where the solute concentration in solid and liquid are uniform throughout the phase. The diffusivity of solute particles is usually several orders higher in liquid state than in solid. Moreover, liquid portion is also subjected to advection through forced (stirring, flow field etc.) or natural causes (shrinkage). Thus, liquid concentration within the domain under consideration is homogenized instantly. Solid concentration however, needs special attention as the characteristic diffusion time is typically very large as compared to the time scale for solidification.

The most common means of representing the role of solute concentration in governing phase change is through the equilibrium phase diagrams and the classical Lever Rule in terms of the solid fraction, $g$, as:

$$C_L = \frac{C_o}{1 - (1 - k_p)g} \quad (2.29)$$

However, application of the lever rule is restricted to cases undergoing infinitely slow cooling. The solute deposited in solid state has enough time to attain the state of equilibrium so that the concentration distribution is uniform. Infinite solid state diffusivity is
a characteristic assumption of the lever rule. The theory is widely used to study formation of microstructures in large castings and directional solidification where the cooling rates are externally controlled. Isomorphous systems can be accurately analyzed with this approach. However, certain eutectic systems do deviate from the lever rule even at slower cooling rates. To understand this situation, consider an initial alloy composition of \( C_0 < C_\alpha \). Through the application of Equation 2.29, it can be seen that the solid concentration can never reach \( C_{\text{eut}} \). Contrary to this prediction, microstructural observations in certain situations do exhibit a lamellar structure which is characteristic of a eutectic solidification, thereby violating the equilibrium assumption. Eutectic formation can be explained by non-equilibrium solidification theories proposed in the literature [31] where a finite solid state diffusivity is accounted for.

The rapid solidification model [31], also called Scheil-Gulliver theory is the other extreme of non-equilibrium phase change where the solid state diffusivity is completely neglected. Under such assumption, the following equation can be derived to relate the solidification process with the concentration.

\[
C_L = C_\alpha (1 - g)^{k \rho - 1}
\] (2.30)

An important conclusion of this theory is that a eutectic microstructure is always predicted, irrespective of the initial concentration. The eutectic fraction can be obtained by taking \( C = C_{\text{eut}} \) in Equation 2.30. Figure 2-6 schematically explains the process of formation of a single solid grain undergoing infinitesimally slow and rapid solidification along with the concentration profiles.

The formation of a solid phase from a pool of liquid is in the form of grains. Each grain nucleates at discrete locations and undergoes growth dictated by specific non-equilibrium solidification theories. At complete solidification, discrete grains are observed, each having a unique concentration profile. Grain (dendrite) growth is a microscopic
process and its modeling requires phase field models [59]. The present work assumes a volume averaging technique to model dendritic growth throughout the domain. A single representative concentration profile is assumed and the solute conservation equation is formulated. A schematic of a general concentration profile is given in Figure 2-7.

![Equilibrium and Rapid Solidification Schematics](image)

**Figure 2-6 Equilibrium and Rapid Solidification Schematics**

![General Concentration Profile](image)

**Figure 2-7 General Concentration Profile**

The concentration gradient between the center of the grain and the surface drives the diffusion of solute particles in the solid state. This process is called Back
Diffusion. It can easily be inferred that zero back diffusion corresponds to rapid solidification while complete back diffusion represents a uniform profile corresponding to the lever rule. Other non-equilibrium solidification theories lead to a specific concentration profile based on fundamental assumptions. The first attempt to account for back diffusion was by Brody and Flemmings [29]. A constant growth rate was assumed to derive a closed form solution for the concentration profile under the combined effects of solute partitioning and back diffusion. The mathematical representation of this model could be written as:

\[ C_L = C_o \left( 1 - \frac{g}{1 + \alpha_c k_p} \right)^{k_p^{-1}} \]  

(2.31)

where \( \alpha_c = 4D_s t_{sol}/l^2 \) is the called the dimensionless diffusivity or the Fourier number. \( D_s \) is the mass diffusivity of solute particles in solid phase. The length, \( l \), is typically taken as half the grain size or the interdendritic spacing and \( t_{sol} \) is the solidification time. Equation 2.31 can be viewed as a modification of the Scheil-Gulliver theory. It is observed from the analysis that this model provides good accuracy for low values of diffusivity only. Attempts to solve this problem were made by many researches. Two of the most significant modifications made to this formulation were from Clyne and Kurz [27] and Ohnaka [28]. Both these modifications were based on empirical relationships for solute concentration profiles in solid state. These theories are summarized in Table 2-1 following a discussion on modeling back diffusion in the present work.

2.4.3 Solute Conservation

Dendritic growth in alloys is a micro-scale phenomenon. Hence, analysis of such a process along with modeling of macroscopic quantities like temperature and velocities poses a mathematical challenge. Ni and Beckerman [34] suggested an approach of
modeling macroscopic quantities as volume average of the corresponding microscopic quantities on a scale as small as that of the half arm spacing in dendrites. Such a scale ensures that complete information is accounted for in the volume averaging. These quantities can be taken as the representative of all the dendritic scales involved in a macroscopic computational cell. It makes the numerical formulation and analysis involving micro-scale phenomenon like solute partitioning and back diffusion possible. Figure 2-8 schematically represents an example of REV in the half arm space between two secondary dendritic arms.

![Figure 2-8 The Concept of REV](image)

The thermodynamic quantities are first categorized as macroscopic and microscopic quantities. Temperature and velocity are considered macroscopic which means that these quantities do not change within the length scale of a REV. Concentration is a microscopic quantity. If \( g = s/X \) defines the solid fraction in a REV, the overall solute concentration is given by:

\[
C = g\bar{C}_s + (1 - g)C_l
\]

(2.32)

where \( \bar{C}_s \) is the average solid concentration and \( C_l \) is the liquid concentration which is uniform. Determination of the changes in average solid concentration forms the basis of this formulation. Average solid concentration can be written as

\[
\bar{C}_s(g) = \frac{1}{g} \int_0^g C_s(f) \, df
\]

(2.33)
The general transport equation for solute concentration can be written as

$$\frac{\partial C}{\partial t} + \nabla \cdot (VC) - \nabla \cdot (D \nabla C) = 0$$

(2.34)

Where V is the liquid velocity and D is the macroscopic solute diffusivity. In the present study it has been assumed that the solid portion remains stationary while liquid motion is treated as one in a pseudo-porous medium. Therefore the advection and diffusion terms will only be applied to liquid concentration. Using Equation 2.32 in Equation 2.34 gives

$$\frac{\partial}{\partial t} (g \bar{C}_s + (1 - g) C_l) + V \cdot (V(1 - g)C_l) - \nabla \cdot (D \nabla (1 - g)C_l) = 0$$

(2.35)

Applying chain rule to the first term,

$$\frac{\partial}{\partial t} (g \bar{C}_s) = \int_0^g \frac{\partial}{\partial t} [C_s(f)] df + \bar{C}_s \frac{\partial g}{\partial t}$$

(2.36)

It has to be noted that the time derivative of the solid concentration requires information about $C_s(g)$ which can be derived from the specific non-equilibrium theory mentioned earlier. However, it is practically inappropriate to consider one specific approach as each has fundamental limitations pertaining to the cooling conditions. Therefore, Voller et al. [37] suggested an approximate back diffusion treatment that can be applied in general without having to consider one specific approach or concentration profile. The rate of change of average solid solute concentration is expressed as

$$\frac{1}{g} \int_0^g \frac{\partial [C_s(f)]}{\partial t} df = \beta \frac{\partial [C_s(g)]}{\partial t}$$

(2.37)

where $\beta$ is called the back diffusion parameter. It takes different values depending on the scheme used for the concentration profile. Table 2-1 enlists the four non-equilibrium solidification theories proposed in the literature with the corresponding suggested values for the back diffusion parameter.
Table 2-1
Summary of Solidification Models

<table>
<thead>
<tr>
<th>Model</th>
<th>Back Diffusion Parameter, $\beta$</th>
<th>Model Assumptions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Scheil-Gulliver [31]</td>
<td>0</td>
<td>No diffusion (Rapid)</td>
</tr>
<tr>
<td>Lever Rule [31]</td>
<td>1</td>
<td>Infinite diffusivity (Equilibrium)</td>
</tr>
<tr>
<td>Brody-Flemings [29]</td>
<td>$2\alpha$</td>
<td>Constant growth rate</td>
</tr>
<tr>
<td>Clyne-Kurz [27]</td>
<td>$2\alpha \left[1 - \exp\left(-\frac{1}{\alpha}\right)\right] - \exp\left(-\frac{1}{2\alpha}\right)$</td>
<td>Exponential concentration profile</td>
</tr>
<tr>
<td>Ohnaka [28]</td>
<td>$\frac{2\alpha}{1 + 2\alpha}$</td>
<td>Quadratic concentration profile</td>
</tr>
</tbody>
</table>

A generalized classical closed form solution using this approximate treatment has been derived as [38]:

$$g = \frac{1}{1 - \beta k_p} \left(1 - \left(\frac{C_1}{C_0}\right)^{1 - \beta k_p}ight) \quad (2.38)$$

The two extreme cases with equilibrium and rapid solidification can be regained from Equation 2.38 by taking $\beta = 1$ and zero respectively. Therefore, $0<\beta<1$ gives different concentration profiles for varying diffusivities. Figure 2-9 compares the classical theories for $C_o = 4.5$ wt % and $k_p = 0.15$. The profiles are compared for two values of Fourier number. Note that the Brody-Femmings equation has certain fundamental limitations. As mentioned earlier, Equation 2.31 is only applicable to when the solid state diffusivity is
considerably small. For larger values of $D_s$ (or $\alpha$), the back diffusion parameter may be greater than unity and result into physically unrealistic predictions. The treatment works better for Clyne-Kurz and Ohnaka models over a wide range of diffusivities. For highly accurate values of back diffusion parameter, the characteristic diffusion time $t_s$ needs to be measured implicitly as suggested in [60]. However, at very high cooling rates as in the case under study, a constant back diffusion parameter [37, 38] yields satisfactory results.

Figure 2-9 Comparison of Concentration Profiles
The above discussion is based on the assumption of a fixed scale of an REV. In reality, as the solidification progresses, the dendritic arm spacing widens as shown in Figure 2-10. The arm scale is empirically related as $X \alpha \tau^n$ where $\tau$ is the dimensionless time and $n$ is the coarsening exponent which is experimentally determined to be $1/3$ [61].

Actual treatment for arm coarsening effect needs microscopic modeling methods. The coarsening of the half arm space or the REV leads to dilution of the liquid in that particular REV. Therefore, Voller and Beckerman [58] showed that coarsening can be combined with back diffusion to make the mathematical modeling simpler. The Fourier number is modified as:

$$\alpha^+ = \alpha + \alpha^c$$  \hspace{1cm} (2.39)

A wide range of study has been conducted in determining the value of $\alpha^c$ [38, 58, 61]. The following approximation was proposed:

$$\alpha^c = \frac{n}{m + 1}$$  \hspace{1cm} (2.40)

‘m’ is a constant describing the particular concentration profile assumption. Satisfactory test results have been reported for $m = 2.33$ which gives $\alpha^c = 0.1$. This correction
parameter is applicable for a wide range of materials and process conditions. A brief discussion regarding the effects of arm coarsening has been presented in later sections. However, these effects are more evident at low cooling rates such as ones experienced in directional solidifications and solidification in molds.

Including the approximate back diffusion treatment in Equation 2.35, the solute transport equation can be re-written as

\[
\frac{\partial}{\partial t} (g\overline{C_s}+(1-g)\overline{C_i}) = \left((1-g)+\beta g k\right) \frac{\partial C_i}{\partial t} - (1-k)\frac{\partial g}{\partial t}
\]  

(2.41)

Rearranging the above equation to make it suitable for numerical discretization, we get

\[
\left((1-g)+\beta g k\right) \frac{\partial C_i}{\partial t} + \nabla \cdot (V(1-g)C_i) - \nabla \cdot (D\nabla (1-g)C_i) = S_s
\]  

(2.42)

where

\[
S_s = (1-k)C_i \frac{\partial g}{\partial t}
\]  

(2.43)

is the solute source term contributing the effects of solidification analogous to Equation 2.18. It has to be noted that no assumptions regarding grain morphology have been made so far which means that the formulation is applicable to isomorphous as well as eutectic systems. Solution of the governing equations for isomorphous systems is easier because of a continuous single phase growth. Eutectics have a discontinuity at a specific concentration which needs further consideration. Although, modeling the solidus and liquidus curves in isomorphous systems, i.e. \(T_{equ} = f(C_i)\) in the equilibrium phase diagram might be non-linear, needing further simplifications.

2.4.4 Treatment for Eutectic Solidification

The scheme outlined in section 2.4.3 is only valid above the eutectic temperature. The peculiar eutectic solidification behavior makes it difficult to model
further solidification. Voller [25] suggested extrapolating the liquidus line beyond eutectic to enable the model attain complete solidification. This approach clearly violates the physics of eutectic growth. The classical non-equilibrium solidification theories also fail to predict complete solidification. For example, the Scheil equation gives physically unrealistic concentration values as \( g \rightarrow 1 \). Therefore, numerical modeling of the complete solidification process requires special focus. The method proposed here facilitates the simulations to continue until complete solidification without the need for extrapolating the liquidus line. Such a treatment is needed to study the exact locations and distribution of eutectic throughout the domain. Moreover, the overall cooling characteristics, spreading and solidification characteristics and temperature profiles require complete analysis of the phase change process.

Consider the solidification case as shown in Figure 2-11. \( C_0 \) is the initial composition of the molten mixture. After a period of time the solidification process will reach the eutectic point. It is assumed that no solute separation takes place in a very small range of temperature ‘\( \delta \)’ below eutectic. This assumption is appropriate considering the objective of the present study. The new solid deposited below eutectic temperature is lamellar and can be treated as a homogeneous mixture of ‘\( \alpha \)’ and ‘\( \beta \)’ phases while the already formed dendritic ‘\( \alpha \)’ phase remains undisturbed. Any further change in solid fraction is due to lamellar deposition alone. Therefore a model similar to the one described in section 2.4.1 is employed for eutectic solidification. The fraction of liquid remaining when \( C = C_{\text{eut}} \) is

\[
\theta_{\text{eut}} = (1 - g)_{C_l = C_{\text{eut}}}
\]

which is called the eutectic fraction. The temperature in the mushy region below eutectic is expressed as a linear function of liquid fraction as represented below.
\[ \theta = \theta_{eut} \left( \frac{T - T_{eut} + \delta}{\delta} \right) \] (2.45)

Such a treatment ensures that accurate eutectic prediction is retained in the scheme as it is done through the non-equilibrium solidification principles.

Figure 2-11 Special Treatment for Eutectic Solidification
Chapter 3
Numerical Model

3.1 Flow Solver

The flow solver is based on the works of [53, 55, 62]. The traditional Navier-Stokes and continuity equations are modified to accommodate the volume averaged fluid velocity as [24]

\[ \nabla \cdot \bar{\mathbf{v}} = 0 \]  
\[ \frac{\partial \bar{\mathbf{v}}}{\partial t} + \nabla \cdot (\bar{\rho} \bar{\mathbf{v}}) = -\frac{\partial p}{\partial t} + \bar{\mathbf{f}} + \frac{\partial \bar{\mathbf{g}}}{\partial t} + \bar{\mathbf{f}}_b + \mathbf{S}_v \]  

A two-step projection algorithm outlined in [62] is used to solve for the flow field. Equation 3.2 is discretized in time as

\[
\frac{\bar{\mathbf{v}}^{n+1} - \bar{\mathbf{v}}^n}{\Delta t} = - \nabla \cdot (\bar{\mathbf{v}}^{n+1}) - \frac{\partial n}{\partial t} \nabla p^{n+1} + \bar{\mathbf{f}}^{n} + \bar{\mathbf{g}}^{n} + \bar{\mathbf{f}}_b^{n} + \mathbf{S}_v^{n}
\]  

where \( \mathbf{S}_v^{n} \) represents an intermediate velocity. In the first step, \( \bar{\mathbf{v}}^{i} \) is computed from Equation 3.4 which accounts for the incremental changes resulting from viscosity, advection, gravity, and body forces (\( \mathbf{F}_b \) and \( \mathbf{S}_v \)). The second step involves taking the divergence of Equation (16) while projecting the velocity field, \( (\bar{\mathbf{v}})_{n+1}^{n+1} \) onto a zero-
divergence vector field for mass conservation. This results in a single Poisson equation for the pressure field given by:

$$\frac{\nabla \cdot \vec{V}}{\delta t} = \nabla \cdot \left[ \frac{\theta^n}{\rho^n} \nabla P^{n+1} \right]$$

(3.6)

Equation 3.6 is solved using an Incomplete Cholesky Conjugate Gradient technique (ICCG) [63]. It is important to note that the density term $\rho^n$ is kept inside the divergence operation even when an incompressible fluid is considered. This is done to maintain continuity across the transition region representing interfaces or free surfaces.

The viscous stress components are explicitly calculated. The diagonal elements in the stress tensor are located at the cell centers while the off-diagonal components are located at the cell vertices as shown in Figure 3-1.

![Figure 3-1 Velocity and Viscous Stress Components](image)
Such an arrangement facilitates a straightforward finite difference approximation to compute the respective velocity gradients as

\[ \tau_{xx} = 2\mu \left( \frac{u_{i+\frac{1}{2},j} - u_{i-\frac{1}{2},j}}{\delta x_i} \right)^o \]  

(3.7)

\[ \tau_{yy} = 2\mu \left( \frac{v_{i,j+\frac{1}{2}} - v_{i,j-\frac{1}{2}}}{\delta y_j} \right)^o \]  

(3.8)

\[ \tau_{xy} = \tau_{yx} = \mu \left[ \frac{v_{i+1,j+\frac{1}{2}} - v_{i,j+\frac{1}{2}}}{\delta x_{i+\frac{1}{2}}} + \frac{u_{i+\frac{1}{2},j+1} - u_{i+\frac{1}{2},j}}{\delta y_{j+\frac{1}{2}}} \right]^o \]  

(3.9)

The explicit consideration imposes a stability criterion given by [62]:

\[ \delta t < \frac{1}{3\vartheta} \left( \frac{(\delta x)^2 (\delta y)^2}{(\delta x)^2 + (\delta y)^2} \right) \]  

(3.10)

In case of a non-uniform mesh, the minimum value of the right hand side of the inequality is considered.

Surface tension is evaluated using a continuum surface force method [42]. Surface tension being a surface phenomenon, poses a discontinuity in the governing equations when a free surface is present within the domain. The CSF method interprets surface tension as a continuous three dimensional effect across a small transition thickness near the interface. In other words, interfaces between fluids of different properties are represented as transition regions of finite thickness. Across this region there is a continuous variation from property value of one fluid to the property value of the other fluid. At each point in the transition region, a force density (i.e. body force per unit volume) is defined which is proportional to the curvature of the surface at that point. The conventional description of surface tension on an interface is recovered when the ratio of
local transition region thickness to local radius of curvature approaches zero. This body force is mathematically represented as:

\[
\vec{F}_b = \vec{F}_{sv} = \sigma K \delta(\vec{x}) \hat{n}
\] (3.11)

where \( K \) is the curvature, \( \delta \) is a smoothed delta function and \( \hat{n} \) is the unit vector perpendicular to the surface area. The normal vector in the original CSF method is obtained as a gradient of color function. Color function is a unique quantity used to identify different phases in a domain. In the present formulation, color function is equivalent to the VOF function. Therefore, the unit normal vector is calculated from Equation 2.13 or 2.15 and the curvature is defined as

\[
K = -\nabla \cdot \hat{n}
\] (3.12)

3.2 Interface Tracking and Reconstruction

The fluid free surface is tracked and reconstructed using two volume advection schemes: VOF-PLIC [49] and CLSVOF [53]. The basic volume tracking idea of Hirt and Nicholes [48] is incorporated in both the algorithms. However, the orientation of the interface is calculated with different approaches. The VOF and LS functions defined earlier are first advected under the effect of the flow field using Equation 2.8. Numerical solution of these equations is obtained using the same two-step projection method that was implemented for solving the NS equation.

\[
\frac{\tilde{C} - C^n}{\delta t} + \frac{\partial}{\partial x}(uC^n) = \frac{\partial \tilde{C}}{\partial x}
\] (3.13)

and

\[
\frac{C^{n+1} - \tilde{C}}{\delta t} + \frac{\partial}{\partial y}(v\tilde{C}) = C^{n+1} \frac{\partial v}{\partial y}
\] (3.14)
represent the numerical method for advection of a color function 'C' which takes the value of F for VOF advection and \( \varphi \) for LS advection. Once advected, the LS function loses its definition of being a distance function and needs to be re-initialized. This is done through the process outlined by [55] which uses a normal propagating velocity to correct the LS functions after each advection routine. Although, it must be noted that advection – redistancing sequence will not guarantee mass conservation. Therefore LS and VOF methods are used in conjunction in the CLSVOF scheme. Depending on the choice of the interface tracking method, the unit normal vector is calculated using Equation 2.13 or 2.15. This normal vector is used in the CSF method to calculate surface curvature and also in the interface reconstruction process.

A piecewise linear interface construction scheme is adopted to reconstruct the interface from the VOF functions. The normal vector obtained in the previous step gives the orientation of the free surface. The interface is approximated as a straight line orthogonal to the normal vector as shown in Figure 2-2. Such a treatment has superior free surface definition even when predicting sharp corners. The previous methods which were based on a donor-acceptor algorithm used a SLIC [48, 62] scheme to represent fluid interfaces as either horizontal or vertical lines. Interface curvature calculations were substantially inferior as compared to Young's PLIC scheme [54]. The flux calculations outlined in [49] are implemented in the present work. Moreover, the weighted combinations of upwind and downwind schemes for calculating face fluxes in [62] lead to erroneous diffusion (false diffusion). The concept of a non-zero finite slope of the interface aids in the correction of these terms. The four possible configurations for the interface reconstruction using a PLIC scheme are shown in Figure 3-2. Note that there are sixteen total possibilities. However, the cell rotation and indexing method used by Rudmann [49] reduces the total possibilities to four. The wetting ratios obtained as a
result of interface construction are used to calculate the accurate momentum and thermal fluxes through the respective faces.

Figure 3-2 PLIC Configurations

3.3 Energy and Solute Transport Equations

A staggered grid arrangement shown in Figure 3-3 is employed to discretize the energy and solute transport equations.

Figure 3-3 Staggered Grid Arrangement
The generalized formulation outlined by Patankar [64] is followed with special treatment for the source term. Integration of Equation 2.21 over a computational cell (control volume) yields:

$$\frac{\partial (\rho h)}{\partial t} + \frac{1}{r} \frac{\partial J_r}{\partial r} + \frac{\partial J_z}{\partial z} + S_h = 0,$$  \hspace{1cm} (3.15)

where 'J' represents the net flux through a cell face as:

$$J_r = r \left( \rho u_r h - \alpha \frac{\partial h}{\partial r} \right); J_z = \left( \rho u_z h - \frac{\partial h}{\partial z} \right)$$  \hspace{1cm} (3.16)

and

$$S_h = \frac{\partial (\rho \delta h)}{\partial t} + \frac{1}{r} \frac{\partial (r \rho u_r \delta h)}{\partial r} + \frac{\partial (\rho u_z \delta h)}{\partial z}$$  \hspace{1cm} (3.17)

Integrating the above equation over a control volume by following the notation given in Figure 3-3, the energy equation converts into

$$\frac{(\rho_p h_p - \rho_p^0 h_p^0)\Delta r \Delta z}{\Delta t} + \frac{1}{r} [J_e - J_w] + (J_n - J_s) + S_h \Delta r \Delta z = 0$$  \hspace{1cm} (3.18)

The continuity equation integrated over the control volume yields

$$\frac{(\rho_p - \rho_p^0)\Delta r \Delta z}{\Delta t} + \frac{1}{r} [(\rho u)_e - (\rho u)_w] + [(\rho v)_n - (\rho v)_s] = 0$$  \hspace{1cm} (3.19)

Multiplying Equation 3.19 with $h_p$ and subtracting from Equation 3.18 gives:

$$\frac{\rho_p (h_p - h_p^0) \Delta r \Delta z}{\Delta t} + \frac{\Delta z}{r_p} \left[ (J_e - r_e (\rho u_e)_e h_e) - (J_w - r_w (\rho u_w)_w h_w) \right]$$

$$+ \Delta r [(J_n - (\rho u_n)_n h_n) - (J_s - (\rho u_s)_s h_s)] + S_h \Delta r \Delta z = 0$$  \hspace{1cm} (3.20)

Using the generalized formulation, Equation 3.20 can be written in algebraic form as

$$a_p h_p = a_e h_e + a_w h_w + a_n h_n + a_s h_s + a_p^0 h_p^0 + b$$  \hspace{1cm} (3.21)

with the coefficients given by Equation 3.22. Note that an upwind advection scheme is used to discretize the source term. Introduction of certain degree of underrelaxation may be necessary depending on the thermodynamic properties to ensure stability.
\[ a_e = D_e A(|P_e|) + \| -F_e, 0\| \]
\[ a_w = D_w A(|P_w|) + \| F_w, 0\| \]
\[ a_n = D_n A(|P_n|) + \| -F_n, 0\| \]
\[ a_s = D_s A(|P_s|) + \| F_s, 0\| \]
\[ a_p^o = \frac{\rho_p^2 2\pi r^2 \Delta r \Delta z}{\Delta t} \]
\[ a_p = a_e + a_w + a_n + a_s + a_p^o \quad (3.22) \]

\[ b = a_p^o (\delta h_p - \delta h_p^0) + \text{inflow} - \text{outflow} \]

\[ \text{outflow} = [\| F_n, 0\| (\delta h)_{p} - \| -F_n, 0\| (\delta h)_{n}] \]
\[ + [\| F_e, 0\| (\delta h)_{p} - \| -F_n, 0\| (\delta h)_{e}] \]

\[ \text{inflow} = [\| -F_s, 0\| (\delta h)_{p} - \| F_s, 0\| (\delta h)_{s}] \]
\[ + [\| -F_w, 0\| (\delta h)_{p} - \| F_w, 0\| (\delta h)_{w}] \]

where

\[ D_e = \frac{2\pi r_e \alpha \Delta z}{(\Delta r)_e} \quad F_e = 2\pi r_e (\rho u_e) \Delta r \Delta z \quad P_e = \frac{F_e}{D_e} \]
\[ D_w = \frac{2\pi r_w \alpha \Delta z}{(\Delta r)_w} \quad F_w = 2\pi r_w (\rho u_w) \Delta r \Delta z \quad P_w = \frac{F_w}{D_w} \]
\[ D_n = \frac{2\pi r_n \alpha \Delta r}{(\Delta z)_n} \quad F_n = 2\pi r_n (\rho u_n) \Delta r \Delta z \quad P_n = \frac{F_n}{D_n} \]
\[ D_s = \frac{2\pi r_s \alpha \Delta r}{(\Delta r)_s} \quad F_s = 2\pi r_s (\rho u_s) \Delta r \Delta z \quad P_s = \frac{F_s}{D_s} \]

The solute transport equation is discretized following a similar approach. The final algebraic equation can be written as

\[ a_{pc} C_{tp} = a_{ec} C_{te} + a_{wc} C_{tw} + a_{nc} C_{tn} + a_{sc} C_{ts} + a_{pc}^o C_{tp}^o + b_c \quad (3.23) \]

with the coefficients given by Equation (3.24).
\[ a_e = D_{c,e}A(\|P_{c,e}\|) + \| - F_{c,e}, 0 \| \]
\[ a_w = D_{c,w}A(\|P_{c,w}\|) + \| F_{c,w}, 0 \| \]
\[ a_n = D_{c,n}A(\|P_{c,n}\|) + \| - F_{c,n}, 0 \| \]
\[ a_s = D_{c,s}A(\|P_{c,s}\|) + \| F_{c,s}, 0 \| \]
\[ a_p^0 = \frac{2\pi r_p \Delta r \Delta z}{\Delta t} \]
\[ a_p = a_e + a_w + a_n + a_s + a_p^0 \left[ (1 - g_p^o) + \beta g_p k \right] \]
\[ b_e = a_p^0 \left( 1 - k \right) C_t \left( g - g^o \right) \]

where
\[ D_{c,e} = \frac{2\pi r_e D \Delta z}{(\Delta r)_e} \quad F_{c,e} = 2\pi r_e (u_r)_e \Delta z \quad P_{c,e} = \frac{F_{c,e}}{D_{c,e}} \]
\[ D_{c,w} = \frac{2\pi r_w D \Delta z}{(\Delta r)_w} \quad F_{c,w} = 2\pi r_w (u_r)_w \Delta z \quad P_{c,w} = \frac{F_{c,w}}{D_{c,w}} \]
\[ D_{c,n} = \frac{2\pi r_p D \Delta r}{(\Delta z)_n} \quad F_{c,n} = 2\pi r_p (u_z)_n \Delta r \quad P_{c,n} = \frac{F_{c,n}}{D_{c,n}} \]
\[ D_{c,s} = \frac{2\pi r_p D \Delta r}{(\Delta r)_s} \quad F_{c,s} = 2\pi r_p (u_s)_s \Delta r \quad P_{c,s} = \frac{F_{c,s}}{D_{c,s}} \]

3.3.1 Homogeneous Mushy Region Model

This is a simplified solution procedure based on the assumption of a homogeneous mushy region or a fixed mushy range [24]. Solid, liquid and mushy zones are only characterized by a mathematical function 'θ' defined by Equation 2.26. Equation 3.21 is solved in conjunction with Equation 2.26 and the solute transport equation is completely ignored. The sensible heat at any point within the mushy region can be given by definition as:
\[ h_p^* = \int c_p dT = h_s + \int_0^\theta c_{pm}(T_l - T_s) d\theta \]  

(3.25)

where \( c_{pm} \) is the specific heat in the mushy region. In general, specific heat varies with temperature.

The solution procedure starts with a guess value for the liquid fraction. For simplicity, this initial value is directly taken from the previous time step value. Equation 3.21 can be solved for enthalpy as

\[ h_p = \frac{1}{a_p} \left[ \sum_{nb} a_{nb} h_{nb} + b \right] \]  

(3.26)

With such an initial guess, the resulting enthalpy difference \( (h - h^0) \) would be overestimated as the source term vanishes. The next step is to calculate the equilibrium enthalpy defined by Equation 3.25. Using this in Equation 3.21, the liquid fraction correction equation can be derived as

\[ \theta_p^{n+1} = \theta_p^n + \frac{a_p}{L \cdot a_p^p} (h_p^{n+1} - h_p^*). \]  

(3.27)

This value is then used in Equation 3.26 and the three step iteration procedure given by Equations 3.26, 3.25 and 3.27 continues until the convergence criterion is met. A detailed flow chart showing the algorithm is given in Appendix C. If the material properties are assumed constant, the scheme will converge extremely fast because the enthalpy versus temperature relationship is truly linear. Even with the consideration of temperature dependent thermal diffusivity, the scheme converges significantly fast. Also note that no distinction is made in the governing equations pertaining to the state of matter. The same scheme is applicable throughout the domain. Except within the mushy region, the source term becomes dormant and the conventional energy equation is regained.

This simplified scheme is a very good replacement of the actual coupled solution procedure. Isomorphous alloys can be easily modeled using this procedure and will
require fewer computational resources. The only compromise is that the solute concentration could not be modeled implicitly. Empirical calculations of concentration profiles using the temperature distribution may be employed. On the other hand, it may prove to be too large a compromise when it comes to eutectic behavior. Figure 3-4 shows the expected temperature variations with liquid fraction at constant solidification rate in a pure substance, eutectic and isomorphous systems along with the assumption used here. Clearly, eutectic solidification needs special mention as the behavior is far from the homogeneous mushy region assumption. Detailed model comparison is given in subsequent sections.

![Figure 3-4 Cooling Curves for Different Materials](image)

3.3.2 Coupled Temperature-Solute Model

As evident from Figure 3-4 the role of solute separation and back diffusion in governing phase change in eutectic systems may not be overlooked. Equations 3.21 and 3.23 can be solved simultaneously to obtain an accurate solidification behavior. The
procedure starts in the same manner as described in the previous section. However, focusing on solid fraction rather than liquid fraction is more meaningful considering the form of Equation 3.23. Therefore, \( g = 1 - \theta \) transformation is performed at the beginning of the scheme. Taking an initial guess for the solid fraction, the energy equation is solved for the sensible enthalpy using Equation 3.26. The solute transport is also solved as

\[
C_{tp} = \frac{1}{a_{pc}} \left[ \sum_{nb} a_{nb} C_{tnb} + b_c \right] 
\]

(3.28)

The next step is to identify the corresponding equilibrium temperature from the phase diagram using Equation 2.25. The sensible enthalpy at this temperature is given by

\[
h_p^* = c_p T_p^* = c_p \left( T_f + m_l C_{lp} \right) 
\]

(3.29)

If \( C_{tp} = C_{eut} \), the eutectic point is encountered. The corresponding liquid fraction is recorded and stored as \( \theta_{eut} \). In such a case, the special eutectic technique is employed. The equilibrium enthalpy is obtained in a manner similar to the one given by Equation 3.25 and minor modifications are required to express the value in terms of eutectic enthalpy as

\[
h_p^* = h_{eut} - \int_{\theta}^{\theta_{eut}} c_{pm} \delta d\theta 
\]

(3.30)

Using the appropriate equilibrium enthalpy value in Equation 3.27, the liquid fraction (and solid fraction) is updated and used in Equations 3.26 and 3.28 for subsequent iterations until convergence.

It has to be noted that both the source terms i.e. \( b \) and \( b_c \) are dependent on the extent of phase change. Therefore, both the energy and solute transport equations converge simultaneously. It also indicates that the local equilibrium condition imposed by the phase diagram is satisfied implicitly. Accuracy of the scheme depends on the form of \( A|P| \) and \( A|Pc| \) used in the discretization procedure which is highly problem specific.
Therefore careful consideration must be given to the use of this function. For the present problem, a hybrid scheme is used for the energy equation and an upwind scheme for the solute transport.

The model described here is applicable to all binary. Isomorphous systems do not need any further manipulation and the scheme described by Equation 3.29 is sufficient to simulate the complete solidification process. However, the linear liquidus line approximation may not work and needs alternative simplification. On the other hand, the scheme is not enough for eutectic alloys. The discontinuity in the phase change process at the eutectic point poses further challenge. A special treatment to overcome this issue is discussed in the subsequent section. A general algorithm describing the model is given in Appendix C.

3.4 Boundary Conditions

3.4.1 Thermal Contact Resistance

Boundary conditions do play a decisive part in any numerical solution. Attinger et al. [9] and others [19] lay importance on the consideration of contact resistance in governing the heat transfer to the substrate. The contact effects are microscopic phenomenon and theoretical evaluation is rather difficult. Experimental findings do prove the importance of such considerations. Holt [65] outlined an indirect method to compute the boundary heat transfer by accounting for the contact resistance. Similar approach is also found in the works of [66, 26] which has been adopted in the present model.

Contact resistance is incorporated as a modification to the isothermal boundary condition. Imperfect contact (reduced contact area) and entrapment of non-conducting fluids (voids) result into a reduced heat transfer across the contact surface. The resultant
effect can be viewed as an increase in the interface temperature. To determine the modified temperature, a steady state one dimensional conduction model is used. The equivalent thermal resistance [67] can be written as

\[ R_{th} = \frac{l}{kA} + \frac{1}{h_cA} \]  

(3.31)

where \( A \) is the contact area and \( h_c \) is the approximate heat transfer coefficient representing contact conductance per unit area. Attinger [26] suggested a dimensionless approach for evaluating \( h_c \) through the use of Biot number (\( B_i = hl/k \)). The works of [12, 65, 26] suggest a heat transfer coefficient in the range of \( 10^4 \) to \( 10^7 \) W/m²-K for microdroplets while Attinger and Poulakikos [26] suggest a range of \( Bi = 0.01 \) to 2 for accurate thermal resistance modeling through experimental verification. This recommendation is closely followed in the present work.

Using the equivalent resistance from Equation 3.31, the heat transfer from the droplet to the substrate can be calculated as:

\[ q_b = \frac{T^+ - T^-}{R_{th}} = \frac{T_d - T_{sub}}{l\frac{1}{kA} + \frac{1}{h_cA}} \]  

(3.32)

where \( T^+ \) and \( T^- \) represent the theoretical temperatures above and below the interface which translate into the droplet temperature and the isothermal substrate temperature in the present domain. Using a half control volume approach, the interface temperature can be calculated by equating the one dimensional (axial) fluxes on both sides of the boundary as shown in Figure 3-5.

![Figure 3-5 Thermal Contact Resistance Formulation](image)
\[
\frac{T_{d} - T_{if}}{\left(\frac{\delta y}{2kA}\right)} = \frac{T_{if} - T_{sub}}{\left(\frac{1}{h_cA}\right)}
\]

(3.33)

gives

\[
T_{if} = \frac{T_{d} + \frac{h_c}{k} \left(\frac{\delta y}{2}\right) T_{sub}}{1 + \frac{h_c}{k} \left(\frac{\delta y}{2}\right)}
\]

(3.34)

where \(\frac{h(\delta y)}{k} = Bi_{cel}\) represents the cell Biot number. The verification of this scheme can be found in the works of [66, 68]. Although, it provides accurate results, careful consideration of the thermal contact conductance is needed. Figure 3-6 shows the interface temperatures versus droplet temperature for different cell Biot numbers. It can be inferred that even for very high Biot numbers, i.e. perfect contact, the interface temperature is almost 50 K higher than the substrate temperature which may have a significant effect on the dynamics of the problem.

![Figure 3-6 Interface Temperatures for various Cell Biot Numbers](image)

Figure 3-6 Interface Temperatures for various Cell Biot Numbers
3.4.2 Free Surface

As described in earlier sections, the free surface is constructed using a PLIC scheme which represents the interface as a straight line within a computational cell. The momentum and thermal flux calculations across cell faces containing interfaces need special accounting. Such cell faces are partial and the governing equations are not integrated throughout the complete face. The wetting ratios defined in Figure 3-2 are incorporated in the calculation. Taking the example of Figure 2-2, Equation 3.20 can be modified as

\[
\frac{\rho_p (h_p - h_p^0) \Delta r \Delta z}{\Delta t} 
+ \frac{\Delta z}{r_p} \left[ aw_e (f_e - r_e(\rho u_r)_e h_e) - aw_w (f_w - r_w(\rho u_r)_w h_w) \right] 
+ \Delta r \left[ aw_n (f_n - (\rho u_z)_n h_n) - aw_s (f_s - (\rho u_z)_s h_s) \right] 
+ S_h \Delta r \Delta z = 0
\] (3.35)

with the source term being

\[
b = a_p^0 (\delta h_p - \delta h_p^0) + (in\ flow - out\ flow) - a_p^0 \delta h_p^0 (F_p - F_p^0)
\] (3.36)

where the last term represents the enthalpy correction due to mass inflow or outflow over a particular time step. Equation 3.35 suggests that multiplying the F and D terms in Equation 3.22 is sufficient to account for the partial face fluxes near the free surface. Note that the original definition of all the terms is retained within the fluid where all the wetting ratios are equal to one and the mass flow term vanishes from Equation 3.36 because \( F_p = F_p^0 = 1 \). It is important to note that the convection through the free surface has been neglected in the present work, thereby treating the free surface as adiabatic. The first order heat transfer analysis presented in [65] reveal that free surface convection accounts for less than 1% of the total heat content of the liquid droplet.
Chapter 4  
Model Validation

The five important aspects of the numerical model developed in the present work are listed below.

1. Flow Solver
2. Free Surface Tracking and Reconstruction
3. Enthalpy - Porosity Method
4. Coupled Energy and Solute Transport Formulation

The flow solver and free surface tracking algorithms are inherited from the works of [55, 62]. The reader is directed to those references for more information on accuracy and capabilities of the two-step projection method, VOF-PLIC and CLSVOF schemes described in the previous section. The objective of the present study is to analyze the thermal and phase change behavior in a droplet impingement process. Hence, the main focus of this section is the validation of the enthalpy-porosity method and the coupled temperature – solute model.

4.1 Enthalpy – Porosity Technique

The objective of this particular section is to examine the effectiveness of the coupling between energy equation and the multiphase flow solver and the contact resistance formulation. Previous analytical and numerical models [22] were based on the assumption of instantaneous spreading and thus, neglected the simultaneous spreading and solidification behavior. Attinger et al. [68] performed a series of experiments to evaluate this assumption. Their studies involved spreading dynamics of a pico-liter sized droplet with and without freezing. One of the standard cases involving phase change is
considered here. A solder alloy (Sn-38 wt %Pb) is deposited on an isothermal substrate. A non-isothermal solidification assumption was used to simulate the process. Eutectic solder has a fixed melting point of 456 K. For the present simulation, an artificial mushy range of 2 K was introduced below the eutectic point. A parametric study for an optimum contact resistance was carried out and the results are briefly discussed. The heat transfer coefficient appearing in Equation 3.31 was varied between $10^4$ and $10^8$ and the splat formation was verified against experimental data [68]. The interface temperatures throughout the process at the center of the contact surface are shown in Figure 4-1. A substrate temperature of 408 K was prescribed and the interface temperatures were observed to be approximately 10 to 70 K higher over a range of contact resistance values.

![Figure 4-1 Interface Temperatures for various Heat Transfer Coefficients](image)

The final spat shapes and spread factors also appear to be greatly affected as seen in Figure 4-2 and Figure 4-3. An increase in the heat transfer coefficient reduces the interface to substrate temperature difference and consequently, enhances the rate of heat transfer. Solidification occurs faster and arrests the droplet spread.
The formation of splat using a heat transfer coefficient of $7.5 \times 10^5$ W/m$^2$K is given in Figure 4-4 along with the experimental observations of Attinger et al. [68]. The dimensionless height of the splat at the center line ($h/D_0$) and the spread factor ($D_{\text{splat}}/D_0$) obtained from the present study are 0.475 and 1.64 respectively which are in agreement with the corresponding experimentally reported values of 0.45 and 1.6. Figure 4-5 shows a comprehensive agreement of these quantities.
Figure 4-4 Experimental Verification of Splat Formation
Tian et al. [69] conducted experimental studies to understand the role of boundary conditions and substrate properties in a solder jetting process. A solder droplet was made to impinge on different substrates. The first set of study considered a stainless steel substrate with a fluxed surface. The role of flux coating was to suppress the rate of heat loss to the substrate, thereby producing an adiabatic boundary condition. The effect is reproduced in the present work by reducing the approximate heat transfer coefficient to a very small value. Note that in the limiting case of $h_c = 0$ in Equation 3.34, the adiabatic definition is regained. The wall shear and surface tension were also affected although, these quantities are considered to remain constant in the present numerical analysis. Figure 4-6 shows the splat morphologies obtained numerically and experimentally where no solidification was observed.
The second experiment in the studies of Tian et al. [69] was a solder jetting process involving simultaneous spreading and solidification. A non-fluxed, isothermal substrate was considered. The contact heat transfer coefficient recommended in their work was adopted to generate the simulation given in Figure 4-7. A quantitative comparison is done in Figure 4-8 with respect to the spreading of the droplet. Spread factor is defined as the ratio of instantaneous splat diameter to the initial droplet diameter.
Figure 4-7 Splat Morphology Comparison for an Isothermal Substrate

Figure 4-8 Quantitative Comparison of Droplet Spreading Behavior
4.2 Coupled Temperature – Solute Model

The method described in the previous sections is a generalized formulation that accounts for convection and deforming free surfaces in the computational domain. To highlight the effectiveness of the phase change model described in the present work, a simplified domain is considered as shown in Figure 4-9. The dimensions and material properties adopted here are in accordance with the work of [37]. The domain is initially assumed to be occupied by a molten Al-4.5 wt %Cu alloy. The initial concentration is one that corresponds to the solubility limit of the ‘α’ phase shown in the phase diagram in Figure 4-10. Al-Cu system of alloys is a peculiar example where an intermetallic is formed beyond the eutectic composition. However, the scope of present analysis is restricted to $C < C_{\text{eut}}$. The bottom wall of the domain is maintained at a constant temperature of 621 K and the effects of contact resistance are neglected to compare the numerical results with an exact solution having an isothermal boundary. The partitioning coefficient is calculated assuming straight solidus and liquidus lines.

![Diagram](image)

Figure 4-9 1D Directional Solidification Test
The concentration profiles obtained from the back diffusion formulation discussed earlier are compared with the classical solutions of rapid and equilibrium solidification theories. Figure 4-11 shows the concentration profiles during solidification against the local solid fraction. The Lever Rule and Scheil equation profiles are obtained from Equations 2.29 and 2.30 respectively. Two other cases with back diffusion parameters of 0.2857 and 0.4792 are also considered. The observation helps in understanding the role of back diffusion during solidification. The physical scenarios represented by a unique back diffusion parameter are bounded by the classical solutions of infinite and zero back diffusion limits. Voller et al. [37] used the Ohnaka’s [28] (refer Table 2-1) theory to estimate the back diffusion parameter as $\beta = 0.2857$ corresponding to a solute diffusivity of approximately $D_s = 5 \times 10^{-13} \text{m}^2/\text{s}$ [37]. Moreover, the corrected back diffusion parameter to account for arm coarsening effect through Equation 2.39, as suggested by Voller et al. [37] is $\beta^* = 0.4792$. The differences in theoretical predictions of eutectic fractions are clearly visible.
The results from Figure 4-11 can be shown with respect to a similarity variable given by:

$$\eta = \frac{z}{\sqrt{2kt}}$$

(4.1)

where $z$ is the distance from the chill. Mixture concentration is plotted versus $\eta$ in Figure 4-12. A comparison with the similarity solution method of [30] is also made. Moreover, the primary objective of [38] was to have accurate prediction of eutectic formation. The consideration of arm coarsening treatment improved the model substantially. Eutectic fractions obtained through the present work are compared with the corresponding results reported by [38, 70] in Figure 4-13. It can be concluded from the observations that the effects of dendritic arm scaling may not be overlooked especially in directional cooling at slow rates.
Figure 4-12 Comparison of Similarity Profiles [30]

Figure 4-13 Comparison of Eutectic Fraction Predictions [38]
Similar studies were carried out by Diao and Tsai [71] and Cheng and Stefanescu [72] to study and simulate an inverse segregation problem. The domain setup and parameters are similar to the previous case. The following simulations were performed with no back diffusion, i.e. $\beta = 0$. The propagation of the solidification and liquidus fronts were examined and compared with the findings reported in [71] as shown in Figure 4-14. A good agreement was achieved.

![Figure 4-14 Comparison of Propagation of the Solidus and Liquidus Fronts during Rapid Solidification ($\beta = 0$) [71]](image)

In addition to the front propagation, the thermal and concentration distribution at a particular time instant were also examined and compared. Figure 4-15 shows the temperature profile throughout the domain while concentration profile is given in Figure 4-16 at $t = 180$ s.
Figure 4-15 Temperature Profile Comparison for the Inverse Segregation Test [71]

Figure 4-16 Concentration Profile Comparison for the Inverse Segregation Test [71]
The classical Scheil profile for solid fraction, liquid and mixture concentrations are explained in Figure 4-17.

Figure 4-17 Comparison of Species Concentrations and Phase Fractions [71]
Chapter 5
Results and Discussion

Droplet deposition is a process with extensive industrial applications in the area of microscale manufacturing. Optimization of the process parameters require detailed understanding of the physics governing the process. As mentioned in earlier sections, a number of factors such as convection effects, substrate remelting, solidification and droplet deformation have a significant effect on the final outcome. The objective of the present study is to have detailed information on these aspects. The results presented here are broadly classified into two sets. The first set of analyses is associated with an impingement and solidification of a SS304 droplet on a SS304 substrate through the use of the homogeneous mushy region model described above. The primary aim of this study was to examine substrate remelting and the effects of the highly non-linear flow field and consequently convection terms in the energy equation. The results are compared with experimental findings and a good agreement is achieved. A non-intuitive thermodynamic behavior observed through experiments is also addressed. Although widely accepted, the homogeneous mushy region assumption fails to account for certain microscopic phenomenon governing phase transition in alloys. The second set of study corresponds to the application of a coupled temperature-solute model to simulate solidification in a binary eutectic type of solder alloy. This study is primarily focused on exposing the limitations of the simplified approach used earlier. Moreover, it also facilitates the estimation of various microstructural properties which is otherwise not possible. A comparison with classical solutions is also made to evaluate the role of convection in the coupled governing equations. The problem set up, initial and boundary conditions and
thermo-fluid properties and parameters are described below followed by the results and discussion.

5.1 Problem Setup

A schematic of the general droplet deposition process is given in Figure 5-1. One of the important operating parameters in these studies is the nozzle height. The effect of droplet free fall is manifested in the form of impact velocity which is the velocity of the droplet immediately prior to contact with the substrate. It is assumed that all the potential energy of the droplet generated at the nozzle is converted into kinetic energy with negligible in-flight resistance. Typically processes such as solder jetting or solder bumping are carried out with the nozzle being within 25 cm from the substrate and the energy losses are indeed insignificant. Coating processes such as thermal sputtering which require extremely high impact velocities are performed in a controlled environment, typically a vapor chamber to limit energy losses. Recent developments in the field of 3D printing using metals and alloys involve the use of a liquid environment [73] where viscous resistance of the surrounding fluid needs to be accounted for.

Figure 5-1 Schematic of a Droplet Deposition Process
In the present work, the droplet is initially located right above the substrate with the impact velocity calculated using the free fall equation given by:

\[ V_i = \sqrt{2gH_{\text{fall}}} \]  \hspace{1cm} (5.1)

The computational domain setups for both studies are shown in Figure 5-2 and Figure 5-3 with the corresponding boundary and initial conditions given.

Note that the solid substrate in Figure 5-2 is prescribed using a fluid free surface definition and setting the temperature to a value less than the solidus and consequently, the liquid fraction to zero. Distinction between solid and liquid portions are only made through the interpretation of liquid fraction as explained in Equation 2.20. The thickness
of the substrate is large enough so that the adiabatic boundary condition at the bottom wall is a good representation. As evident, droplet deposition can be considered axisymmetric for which the left hand boundary is made a free slip and zero radial temperature gradient wall. The other two boundaries become totally irrelevant in the present analysis. The initial condition in both parts of the study are identical. Part two of the study requires an additional initial condition for solute concentration in the molten alloy droplet which is set to a uniform value throughout the droplet.

Figure 5-3 Computational Domain for Sn-Pb Droplet Deposition Studies

The prescription of an initial free surface is done by assigning a VOF function value. A detailed initial VOF distribution for a part of the domain is given in Figure 5-4. Note that the F = 1 represents cells filled with material, rather than fluid as has been used in classical VOF methods. Calculation of interface VOF is done through a conic function representing a spherical droplet free surface and a linear profile for substrate surface (only for the first set for study).
The material properties for SS304 and Sn-Pb alloy are given in Table 5-1. Note that the specific heat for SS304 is a linear function of temperature while that of Sn-Pb alloy is assumed constant. The equilibrium phase diagram for Sn-Pb alloy is given in Figure 5-5 to depict the eutectic behavior. The assumption made in Equation 2.25 is reflected in the straight liquidus and solidus lines.
Table 5-1
Material Properties used in the Present Study

<table>
<thead>
<tr>
<th>Property</th>
<th>SS304 [23]</th>
<th>Sn- 10 wt %Pb [74]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal Conductivity, $k$ (W/m²K)</td>
<td>18</td>
<td>55</td>
</tr>
<tr>
<td>Density, $\rho$ (kg/m³)</td>
<td>7900</td>
<td>7000</td>
</tr>
<tr>
<td>Dynamic Viscosity, $\mu$ (mPa-s)</td>
<td>4.7</td>
<td>2.2</td>
</tr>
<tr>
<td>Surface Tension, $\sigma$ (N/m)</td>
<td>0.15</td>
<td>0.566</td>
</tr>
<tr>
<td>Contact Angle, $\theta_c$ (°)</td>
<td>90 (Assumed)</td>
<td>135</td>
</tr>
<tr>
<td>Specific Heat $C_p$ (J/kg-K)</td>
<td>445 + 0.16 T (° C) for T &lt; $T_s$ 260</td>
<td></td>
</tr>
<tr>
<td></td>
<td>670 for T ≥ $T_s$</td>
<td></td>
</tr>
<tr>
<td>Solidus Temperature, $T_S$ (K)</td>
<td>1400</td>
<td>-</td>
</tr>
<tr>
<td>Liquidus Temperature, $T_L$ (K)</td>
<td>1460</td>
<td>-</td>
</tr>
<tr>
<td>Species Composition, $C_o$ (wt %)</td>
<td>-</td>
<td>10 wt %Pb</td>
</tr>
<tr>
<td>Fusion Temperature, $T_{fus}$ (K)</td>
<td>-</td>
<td>505</td>
</tr>
<tr>
<td>Eutectic Temperature, $T_{eut}$ (K)</td>
<td>-</td>
<td>456</td>
</tr>
<tr>
<td>Partitioning Coefficient, $k_p$ (-)</td>
<td>-</td>
<td>0.0656</td>
</tr>
<tr>
<td>Back Diffusion Parameter, $\beta$ (-)</td>
<td>-</td>
<td>0 (Scheil)</td>
</tr>
</tbody>
</table>
5.2 Application of a Homogeneous Mushy Region Model

The case of a SS304 droplet impacting, spreading and solidifying with substrate remelting is analyzed with particular focus on the effects of flow field within the droplet. An augmented diffusion limit model was used to study the process numerically by Schmaltz [22, 23] with the objective of indirectly accounting for the convection effects without actually solving for the flow field. A thermal conductivity multiplier called K-factor was approximated by comparing the convective and conduction heat flux across the thermal boundary layer assuming droplet deposition as a flow over a flat plate to artificially enhance the thermal diffusion. K-factor in the range of 2 to 5 was warranted for most cases. In their numerical studies, spreading of the droplet was assumed to occur instantaneously and a predefined fixed shape of the deposited droplet was prescribed in the domain. Such and assumption completely eliminated the involvement of the flow field.
in the process although, it came with serious consequences. A comparison of results obtained by the augmented diffusion limit model and the present study is done to evaluate the consequences of these assumptions and illustrate the role of convection in the energy equation. Experimental findings reported in [22, 23] are used for validation.

A non-uniform quadratically coarsening mesh was used in the present analysis. Further details regarding the mesh and grid refinement test are provided in the Appendix. A standard case with the parameters specified in Table 5-2 was used to compare with the experimental and numerical results from the literature followed by a parametric study to examine the effects of substrate preheating and impact velocity.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial drop diameter</td>
<td>4.3 mm</td>
</tr>
<tr>
<td>Initial drop temperature</td>
<td>2500 °C</td>
</tr>
<tr>
<td>Ambient air temperature</td>
<td>30 °C</td>
</tr>
<tr>
<td>Initial substrate temperature</td>
<td>30 °C</td>
</tr>
<tr>
<td>Impact velocity</td>
<td>0.8 m/s</td>
</tr>
</tbody>
</table>

The numerically obtained remelting depth in the present study is plotted versus time in Figure 5-6 with the corresponding plots from [23] where different k-factors were used. Mathematically, the remelting front is defined as the contour corresponding to θ = 0. As observed from the simulations, the maximum penetration of the mushy region into the substrate occurs at the center. The maximum remelting depth obtained from the
present numerical study is 0.0781 mm while the one experimentally observed is 0.098 mm [23]. It is also evident from Figure 5-6 that the maximum remelting depth varies drastically with the k-factor. An optimum value of K-factor = 5 was proposed in [23] by comparing the remelting depth with experimental measurements. It should be noted that this value leads to inaccurate predictions for other quantities like cooling rates, solidification time and microstructural properties which will be discussed in later sections.

Figure 5-6 Remelting Depth versus Time for an SS304/SS304 Deposition

The movement of the phase change front is governed by the energy balance which is mathematically given as:

\[ L \frac{d\theta}{dt} \bigg|_{T=T_s} = J_m - J_s \]  

(5.2)

where \( J_m \) and \( J_s \) are the instantaneous heat fluxes at the solidification front in the mushy and solid regions respectively. The left hand side of the equation indicates the rate at which the solidus isotherm advances and the right hand side is the net instantaneous
heat flux through the solidification front. Comparison of the two quantities at various instants is shown in Figure 5-7. Remelting ceases when the net flux goes to zero which takes place at 0.05 s. Note that the heat fluxes in Equation 5.2 are solely due to diffusion as the velocities at the solidification front are zero.

![Figure 5-7 Diffusion Flux and Solidification Front Propagation](image)

The temperature variations obtained at the two different locations within the droplet are shown in Figure 5-8. The solidification rate close to the substrate surface is the highest owing to a large temperature gradient whereas the top of the droplet experiences slower cooling. The solidification time measured experimentally is 0.25 s [23] with an overall cooling rate of 4400 °C/s and the one obtained in the present study is 0.235 s at a rate of 4681 °C/s whereas an augmented diffusion limit model yields a substantially slower cooling rate and an extended solidification time as shown in Figure 5-8. Table III gives a detailed comparison. It is evident that augmenting the conductivity fails to accurately address the convective effects on solidification and cooling rates.
Moreover, the predictions are highly sensitive to the value of k-factor which is case specific.

![Figure 5-8 Cooling Curve Comparison](image)

The most critical region in a droplet deposition process is the solid-liquid interface. The strength of this region is determined by the extent of remelting as well as the microstructure of this region which is affected by the rate of solidification. Note that, alloys generally undergo dendritic solidification and the cooling rate plays an important role in the growth process. The nucleation and growth of secondary dendrites is sparse at lower cooling rates which eventually lead to a larger average spacing between the secondary dendrites. The secondary dendritic arm spacing (SDAS) is empirically related to cooling rate during solidification as [75]

$$\lambda = 38 \times T^{-0.44}$$  \hspace{1cm} (5.3)

where $\lambda$ is the SDAS in $\mu$m and $T$ is the cooling rate in °C/s. The cooling rates obtained in the present study and the corresponding SDAS calculated using Equation 5.3 at various
radial locations are shown in Figure 5-9. If diffusion alone was responsible for solidification, the flux and consequently the cooling rates at the droplet-substrate interface would be monotonically decreasing from the periphery to the center. The numerical results and experimental measurements are clearly contradictory to their expectation. The Minimum cooling rate and maximum SDAS is experimentally observed [23] to be in the vicinity of the center which is in agreement with the numerical results shown in Figure 5-9.

Figure 5-9 Instantaneous Cooling Rates and SDAS across the Substrate Surface

In the present study, the flow field and temperature profiles are closely examined to study the physics and role of convection during a droplet deposition process. Figure 5-10 shows the streamlines and isotherms at different time instants. The overall flow process can be categorized into three stages after impact: initial spread, recoil and final spread.
Figure 5-10 Streamlines (Right) and Isotherms (Left) within the Droplet
The initial spread and recoil generate a circulation within the droplet adjacent to the free surface as shown in Figure 5-10. The influence of this flow field is evident in the behavior of the isotherms which are aligned with the streamlines indicating a convection dominant heat transport. The temperature gradients along the substrate surface at various times are shown in Figure 5-11.

![Figure 5-11 Axial Temperature Gradients across the Substrate Surface](image)

For a fixed droplet shape, the thickness is maximum at the center and decreases towards the periphery. Consequently, the temperature gradients are expected to show an inverse trend if the heat equation was solved without the convection terms. Contrary to this prediction, minimum temperature gradient is observed to be in the vicinity of the center. The onset of circulation creates a localized low velocity zone between the central axis and the droplet free surface. Streamlines are pushed away toward the periphery and bypass this zone, effectively shielding it from convective heat transfer. This observation emphasizes the dominance of convection terms in the energy equation. It is impossible to replicate these effects using a diffusion limit method. The basic limitation is the negligence of convection terms in the energy equation which indirectly assumes the
process as being driven by heat diffusion. Although the convection is indirectly accounted for, the fundamental nature of the problem is totally altered resulting in inaccuracies specifically in presence of a non-linear flow field. This peculiar phenomenon indirectly affects the cooling rates, microstructural properties and also the bond strength.

Figure 5-12 shows a comparison of the isotherms and solidification front obtained from the present study, the augmented diffusion limit model and experimental findings [23]. The solidification front predicted from the present study (Figure 5-12 A) resembles the one experimentally observed (Figure 5-12 B) [23]. Moreover, the uniformly varying isotherms in Figure 5-12 C are an outcome of the absence of the convective terms in the energy equation.

![Figure 5-12 Comparison of Solidification Fronts][22, 23]
A comparison of the results obtained from the present study and experimental findings is given in Table 5-3. The overall solidification time predicted from the present study agrees with the experimental finding; whereas the diffusion limit method [23] yields a substantially longer solidification time even at the highest k-factor value of 10. This finding reflects the fact that the diffusion limit method leads to an underestimation of the overall heat transfer when a highly non-uniform flow field prevails. It is also seen that spreading occurs for almost 20% of the solidification time which was overlooked in [22, 23] where a fixed droplet shape was considered.

Table 5-3
Comparison of SS304/SS304 Deposition Results

<table>
<thead>
<tr>
<th></th>
<th>Experimental [23]</th>
<th>Present Study</th>
<th>Diffusion Limit Method [23]</th>
</tr>
</thead>
<tbody>
<tr>
<td>SDAS at the top of droplet</td>
<td>1.09 μm</td>
<td>1.16 μm'</td>
<td>2.07 μm' K = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2.97 μm' K = 10</td>
</tr>
<tr>
<td>Overall cooling rate</td>
<td>4400 °C/s</td>
<td>4681 °C/s</td>
<td>2500 °C/s K = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2619 °C/s K = 2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>2895 °C/s K = 10</td>
</tr>
<tr>
<td>Solidification time</td>
<td>0.25 s</td>
<td>0.235 s</td>
<td>0.44 s K = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.42 s K = 2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.38 s K = 10</td>
</tr>
<tr>
<td>Spreading time</td>
<td>0.05 s</td>
<td>0.08 s</td>
<td>N/A</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>N/A</td>
</tr>
<tr>
<td>Maximum remelting depth</td>
<td>0.098 mm</td>
<td>0.0781 mm</td>
<td>0.0040 mm K = 1</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.0635 mm K = 3.2</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.0947 mm K = 5</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>0.1100 mm K = 10</td>
</tr>
</tbody>
</table>

* Empirical prediction based on numerically obtained value of cooling rate at solidification using Equation 5.3
5.2.1 Effect of Substrate Preheating

Substrates preheated to different temperatures ranging from 150 °C to 300 °C are considered with the rest of the parameters as given in Table 5-2. The cooling rate, spreading, solidification and remelting depth are closely examined.

Figure 5-13 shows the remelting depth for various substrate temperatures. A preheated substrate is already at a higher energy state, thus favoring deeper diffusion of thermal energy and consequently, a higher remelting depth. It can be inferred that with only a finite amount of thermal energy from the droplet, extent of remelting is thermodynamically aided by substrate preheating and that the time to achieve maximum remelting depth is unaffected. Re-solidification, on the other hand, is governed by the total volume of the molten material, thereby taking longer for a preheated substrate. Note that the remelting volume is proportional to the remelting depth because the droplet-substrate contact area is almost the same.
A comparison of the instantaneous cooling rates at solidification for different initial substrate temperatures is shown in Figure 5-14. Preheated substrates have lower cooling rates in general due to a smaller difference between the droplet and substrate temperatures which weakens the rate of heat diffusion.

![Figure 5-14 Cooling Rates with Substrate Preheating](image)

The spreading and solidification times are shown in Figure 5-15. Preheating the substrate has little effect on spreading as previously mentioned because it is primarily governed by the velocity and fluid properties. The solidification times increase marginally for preheated substrates. Thus, the relative period over which spreading occurs reduces with preheating but is significant enough so that the simultaneous effects of spreading and solidification may not be overlooked. Figure 5-16 shows the spread factor versus non-dimensional time which indicates that spreading ceases at around 20% of the solidification time, further emphasizing the argument.
5.2.2 Effect of Impact Velocity

The three impact velocities used for this parametric study are 0.8 m/s, 1.6 m/s and 2.4 m/s with the rest of the parameters as given in Table 5-2. The impact velocities are expected to affect the spreading of the droplet and the contribution of the convection terms in the energy equation.

Substrate remelting for different velocities is shown in Figure 5-17. Higher velocity droplets have a shorter residence time for the heat to diffuse into the substrate.
resulting in a lesser remelting depth for a given substrate temperature. At the same time, the complete phase change process occurs over a longer duration for slower velocities.

Figure 5-17 Remelting Depths for different Impact Velocities

Figure 5-18 shows the cooling rates at the substrate surface for different impact velocities. An increase in cooling rates is observed at greater impact velocities due to a more profound contribution of convection in the heat transfer. Spreading of the droplet on the substrate surface is primarily affected by the impact velocity. It can easily be inferred that a higher impact velocity leads to faster spreading as well as solidification. Moreover, a faster droplet also spreads out farther on the substrate surface. A comparison of the spreading and solidification times is shown in Figure 5-19 and the spread factor is shown in Figure 5-20. Spreading occurs for almost 30 % of the solidification time and is even higher for slower droplets.
Figure 5.18 Cooling Rates for different Impact Velocities

Figure 5.19 Spreading and Solidification Times for different Impact Velocities

Figure 5.20 Spread Factors for different Impact Velocities
5.3 Application of a Coupled Temperature Solute Model

The previous section dealt with a non-isothermal solidification analysis using a homogeneous mushy region assumption. The complete domain was assumed to have a uniform alloy composition at all times. Thus, the role of species concentration in governing phase change was eliminated and the energy equation was solved independently with a particular mathematical approximation. However, modern microscale manufacturing processes such as micro droplet deposition manufacturing (MDDM), thermal sputtering, shape deposition, film coating and solder jetting largely involve the use of eutectic type of alloys. This particular section of the study focuses the thermodynamics, morphology and microstructural properties of a solder jetting process using a Sn-10wt % Pb (solder) molten alloy droplet. Knowledge about the impact dynamics and splat morphology plays a vital role in determining the operational parameters for the particular application. The role of solidification in arresting the spread and thereby controlling the physics of the process is examined. Moreover, the microstructural properties are also estimated empirically based on the numerical results. Parametric studies discussed in the subsequent sections provide thorough understanding of the role of process parameters and material properties on the final outcome.

Studies reported in the literature [17, 19] identify the impact velocity and droplet superheat as two of the most vital governing parameters. Table 5-4 enlists the important non-dimensional parameters and quantities used in the study. The following analysis discusses the effects of these quantities on the morphology and solidification characteristics of the process. The droplet diameter is 1.1 mm and the substrate temperature is 298 K for the reference cases. Reynolds and Stefan number control the
impact conditions in terms of velocity and droplet temperature (superheat) respectively. A grid refinement study is presented in Appendix B.

Table 5-4
Dimensionless Parameters Used in the Present Study

<table>
<thead>
<tr>
<th>Quantity</th>
<th>Definition</th>
<th>Implication</th>
</tr>
</thead>
<tbody>
<tr>
<td>Reynolds Number (Re)</td>
<td>$\frac{\rho V D}{\mu}$</td>
<td>Ratio of inertial to viscous forces</td>
</tr>
<tr>
<td>Stefan Number (Ste)*</td>
<td>$\frac{c_p(T_{id} - T_s)}{L}$</td>
<td>Ratio of sensible to latent heat</td>
</tr>
<tr>
<td>Prandtl Number (Pr)</td>
<td>$\frac{\theta}{\alpha}$</td>
<td>Ratio of momentum to thermal diffusivity</td>
</tr>
<tr>
<td>Weber Number (We)</td>
<td>$\frac{\rho V^2 D}{\sigma}$</td>
<td>Ratio of inertial to surface tension forces</td>
</tr>
<tr>
<td>Spread Factor</td>
<td>$\frac{\text{Splat Diameter}}{\text{Initial Droplet Diameter}}$</td>
<td>Splat diameter</td>
</tr>
<tr>
<td>Dimensionless Thickness</td>
<td>$\frac{\text{Centerline Splat Thickness}}{\text{Initial Droplet Diameter}}$</td>
<td>Splat thickness at center line</td>
</tr>
<tr>
<td>Dimensionless Time</td>
<td>$\frac{\text{Time}}{\text{Solidification Time}}$</td>
<td>Time normalized with solidification time</td>
</tr>
</tbody>
</table>

* Pasandideh-Fard [17] made a distinction on the definition of this dimensionless quantity based on the way the sensible heat is calculated. According to him, Ste represented subcooling normalized with latent heat while Ste represented superheat normalized with latent heat. Ste in the present study will only be used to indicate superheat as defined above.

5.3.1 Splat Morphology and Dynamics

A reference case with Ste = 0.667 and material properties given in Table 5-1 is considered. A set of cases were run with varying Re at the reference Ste value and the solidified splat shapes are compared in Figure 5-21 while the evolution
of splat morphology with flow field and solidification front for two different Reynolds numbers are given in Figure 5-22 and Figure 5-23.

Figure 5-21 Comparison of Splat Shapes for different Impact Conditions

A clear dependence of splat morphology on the impact momentum of the droplet is observed. The extent of spreading is restricted at low impact velocities. The solidification, being a proportional to the contact area is also slower in such circumstances. Complete recoil of the droplet caused by surface tension is observed. At higher impact velocities, a wider spreading and faster solidification arrest the recoil. A monotonous increment in spread factor with impact Reynolds number is observed which indicates that spreading of the splat is directly proportional to the impact momentum of the droplet. An important observation, however, is that the splat shape appears more cylindrical for higher Re impacts which was assumed in some of the previous analytical works [2, 3, 5]. Detailed analysis of this finding and comparison with analytical models will be presented later in the section.
Figure 5-22 Evolution of Splat Morphology for $Re = 3500; Ste = 0.667$
Figure 5-23 Evolution of Splat Morphology for Re = 10500; Ste = 0.667
Instantaneous spread factor for different Re and Ste is given in Figure 5-24 and the same will be compared with analytical results later.

Another important observation made from Figure 5-24 is that there is a saturation in the maximum spread factor which takes place at Re = 14000. Further increase in impact velocity yields in splashing of the droplet. Splashing behavior finds application in film coating processes under very high impact velocities. Extensive work [76] has been reported in the literature on the splashing effects and the formation of satellite droplets under the effect of solidification. The axisymmetric nature of the problem restricts the scope of present analysis to Re < 14000.

The maximum spread factor plays a very important role in dictating the cooling and solidification characteristics of the splat. Figure 5-25 shows a comparison of the maximum spread factor under varying impact Reynolds numbers. The extent of
maximum spread is observed to be approximately 2.8 beyond which splashing occurs. The maximum spread factors for various combinations of Re and Ste are given in Figure 5-26 along with an analytical prediction [17] based on the assumption of negligible energy loss during impact.

![Figure 5-25 Maximum Spread Factor versus Reynolds Number](image)

An expression for the maximum spread factor from the basic energy conservation given as [17]:

\[
\xi_{max} = \frac{We + 12}{\sqrt{\frac{3}{4}Ste + 3(1 - \cos\theta_a) + \frac{4We}{\sqrt{Re}}} + 3(1 - \cos\theta_a) + \frac{4We}{\sqrt{Re}}}
\]

(5.4)

where \(\xi_{max}\) is the maximum spread factor. Comparison with of the terms in the denominator suggest that the effects of solidification can be neglected if \(\sqrt{Ste/Pr} \ll 1\) where Pr = Pe/Re and the classical equation for the spreading of a liquid droplet can be regained. In the cases under consideration, \(\sqrt{Ste/Pr} > 4\) indicating that spreading and solidification cannot be decoupled. Moreover, the observations from Figure 5-26 suggest that the prediction from Equation (5.4 is accurate only at higher Reynolds number. It is
based on the assumption that kinetic energy dissipation is only through solidification while impact losses are neglected. At a low impact velocity, the heat removal through the contact area is considerably small and at the same time the diffusion length scale is very large. Both these effects lead to slower solidification.

However, it is observed from Figure 5-27 that most of the kinetic energy is lost immediately after impact. Thus, even though the thermal energy or solidification dominates the overall phenomenon, the kinetic energy losses during impact are more severe in case of low Reynolds numbers. Moreover, the analytical model did not solve the actual NS equation which means that the actual description of flow field under the effects of surface tension and viscosity and consequently, droplet recoil prevalent at low impact velocity could not be obtained. The formation of splat at high Reynolds number impacts yields a considerably wider spread and faster solidification so that viscous
dissipation and surface tension effects are negligible compared to thermal phenomenon. Figure 5-28 quantitative compares the spread factor obtained from the present study with the corresponding plots given in [4]. The two analytical models are with [17] and without [2] the correction for impact energy losses. It has to be noted that the analytical predictions of Madejski’s model [2] and its derivatives [4] work better at low degrees of superheat because of the inheritance of the Stefan’s result for solidification.

![Figure 5-27 Kinetic Energy Losses during Droplet Deposition on a Substrate](image1)

![Figure 5-28 Comparison of Spread Factor with Analytical Results](image2)
As expected, by increasing $Ste$, the droplet spreads farther out on the substrate exposing more contact area for heat dissipation. Splashing is primarily dependent on the impact dynamics although, increasing the droplet temperature would lead to splashing occurring at relatively lower impact velocities due to localized solidification near the periphery of the splat. It should be noted that although physically realistic, mathematical modeling of contact resistance has a great influence on the computational results. A heat transfer coefficient of $h_c = 4 \times 10^7 \text{ W/m}^2\text{K}$ is found to yield accurate results for the present study and the effects of varying contact resistances will be discussed later. If contact resistance is neglected, solidification starts almost instantaneously irrespective of the impact velocity and results in physically unrealistic splashing behavior. Therefore, careful consideration of contact resistance is warranted for accurate results at high superheats and impact velocities.

Note the transition from 'bump' to 'splat' in Figure 5-21. The transition is observed to be affected by $Ste$ in addition to $Re$. As the droplet undergoes deformation, the inertial momentum is dissipated through two phenomena: viscous dissipation and solidification. Low impact $Re$ typically results in the formation of bump rather than splat. A smaller contact area and a larger droplet thickness result in a slower propagation of the solidification front. Subsequent recoil-spread oscillations are observed which are damped by solidification. On the other hand, high $Re$ impact yields a substantially larger contact area aiding heat removal by the substrate. The solidification is fast enough to dissipate the kinetic energy and arrest the spread and recoil. Splat formation is thus characterized by high impact velocity and thermal inertia. Figure 5-29 and Figure 5-30 depict the phenomenon of bump and splat formation by quantitative comparison of instantaneous droplet thickness observed under various conditions within the domain of the present study. Oscillations in the formation of a bump are clearly visible at the center line, giving a
vivid description. The analytical models [2] were based on decoupled solidification and spreading formulations thereby, failing to account for impact effects and oscillations.

Figure 5-29 Splat Morphologies for Low Reynolds Number Impacts
5.3.2 Solidification and Microstructural Characteristics

Another important aspect in the study of droplet deposition is its solidification characteristics. As mentioned in the above sections, the primitive analytical models were based on the assumption of decoupled solidification and droplet deformation. In other words, impact and initial spread of the droplet were assumed to be unaffected by solidification. Moreover, a simplified solidification description was inherited from the Stefan’s problem to obtain an analytical definition of droplet spread. The total solidification time, front velocity and microstructural developments could not be determined using such assumption. However, microdroplet droplet deposition is used for small scale fabrication processes and determination of accurate thermal as well as flow behavior is of prime importance. A homogeneous mushy region simulates the macroscopic splat profile and overall cooling rates with sufficient accuracy. However,
eutectic alloys require special treatment for the modeling of solidification which is overlooked in the simplified approach. The estimation of material properties requires knowledge regarding the distribution of species composition throughout the solidified part. Microstructural analysis provides valuable assistance in optimizing the process for the desired outcome.

Figure 5-31 shows the overall solidification time at Ste = 0.667 for various Reynolds numbers. Due to the formation of a bump and a very small spread factor, low impact velocity droplets take substantially longer to solidify in comparison to those with a larger contact area. An increased contact area enhances the rate of heat transfer to the substrate in addition to stronger convection effects. As a consequence of the saturation in spread factor shown in Figure 5-25, solidification time is almost constant for large Re impacts.
The solidification times for different Re and Ste are shown in Figure 5-32. It is understood that increasing the thermal inertia of the droplet (i.e. higher Ste) leads to longer solidification times. This is indeed the case at low impact velocities when the spread factor is small. However, greater impact velocities result in almost constant solidification time irrespective of the Stefan number. In such cases, the cooling of the droplet is dictated primarily by the contact area which is almost the same for all Ste. The role of droplet superheat is rendered insignificant.

![Figure 5-32 Overall Solidification Times for different Stefan Numbers](image)

\[ \frac{h_{sol}}{h_{splat}} = \sqrt{\frac{t}{t_{sol}}} \] (5.5)
where $h_{\text{splat}} = U\sqrt{2at_{\text{sol}}}$ is the final splat thickness and $t_{\text{sol}}$ is the solidification time. Note that the above equation is a dimensionless form of Equation 1.2 which is the classical Stefan solution and inherited in [2, 4].

Figure 5-33 Solid Layer Thickness for different Reynolds Numbers

One of the primary objectives of the present work is to analyze the relations between the flow field and solidification characteristics such as thermal and solutal profiles. Impact of the droplet on the substrate has a definitive effect on the temperature profiles within the splat. Figure 5-34 shows the streamlines during the impact and recoil stages and its effect on the corresponding isotherms. The dominance of convection is clearly understood from the thermal behavior especially during impact. The effect of flow field is most exaggerated at the instant of first recoil observed at approximately 2 ms when the isotherms are greatly influenced by the convection flux. Note that the downward flow field results in convex isotherms which is contrary to the expectation of a conduction
governed problem. The diffusion nature of the isotherms is only visible once the flow field subsides. The droplet thickness is maximum at the centerline leading to the lowest temperature gradient and consequently, concave isotherms.

(A) Impact Stage

(B) Recoil and Spread

Figure 5-34 Isotherms (left) and Streamlines (right) for Re = 3500 and Ste = 0.667

Figure 5-35 shows similar observations for the case of Re = 7000. However, due to a very large contact area and a small splat thickness, the effects are not as perceivable as
those at low Reynolds number. Moreover, the solidification occurs fast enough that the convection effects are mitigated within a very short period of time. The solidification front is also almost uniform throughout the splat diameter except for a small region near the circumference where partial recoil is observed. These observations do justify the assumption of a cylindrical splat immediately after impact and are also the reason for near accurate qualitative predictions from the analytical models [4, 17]. Although, the role of convection in governing the heat transfer and solute transport is completely overlooked resulting in inaccuracies.

Figure 5-35 Isotherms (left) and Streamlines (right) for Re = 7000 and Ste = 0.667
Concentration profiles being linearly proportional to temperature, follow a similar trend. Detailed concentration profiles as functions of the local solid fraction across the substrate surface (at 5, 35, 65 and 95 % of the maximum spread) are given in Figure 5-36. The concentration versus solid fraction is expected to follow the Scheil equation (Equation 2.30). However, the strong convection flux from the region of low concentration dominates over the concentration rise due to the solidification source term. Maximum convection flux is observed at 65 % of the spread radius in Figure 5-36 (and also Figure 5-34) directly influencing the local concentration profile. A region in the vicinity of the center of the splat is considered in Figure 5-37 to mimic the stagnation point where the velocities are negligible and the Scheil concentration profile is closely followed.

![Concentration Profiles and Corresponding Momentum Fluxes across the Substrate Surface for Re = 3500 and Ste = 0.667](image)

Figure 5-36 Concentration Profiles and Corresponding Momentum Fluxes across the Substrate Surface for Re = 3500 and Ste = 0.667
To better understand the coupled relation between temperature and concentration, Figure 5-38 shows the solidification period at an arbitrary location in the vicinity of the center of the splat. The relation between temperature and concentration is linear, given by Equation 2.25 and is implicitly satisfied regardless of the rate of solidification or the presence of convection. This fact becomes clear if the results from Figure 5-38 are translated onto a phase diagram in Figure 5-39. The theoretical liquidus line is closely followed without violating the local equilibrium condition imposed by the phase diagram.
The importance of incorporating the coupled phase change model becomes evident when it is compared against the homogeneous mushy region approximation. Figure 5-40 demonstrates the theoretical relation between dimensionless sensible heat and dimensionless latent heat during phase change. The results from a homogeneous mushy region model inherently follow the theoretical assumption given by Equation 2.26. The slope, \( \frac{dT}{d\theta} = 1 \) if the energy equation is decoupled from the solute conservation. However, the coupled model shows significant deviation from this approximation. The slope increases continuously until the eutectic point. A linear profile assumption over a small region of temperature below eutectic is visible in Figure 5-40. Moreover, eutectic composition can easily be identified from this analysis, thus exposing one of the significant limitations of the homogeneous mushy region.
The above analysis demonstrates the fundamental differences between the two models. When determining material properties such as strength, ductility and bonding effectiveness, eutectic distribution within specific regions of the splat provides vital information. Mapping of eutectic throughout the spat is one of the highlighting features of the coupled temperature solute model. Figure 5-41 provides a eutectic map after complete solidification for various combinations of Re and Ste. Microstructure of the splat is characterized by two properties: Secondary Dendritic Arm Spacing (SDAS) for dendritic solidification and Lamellar Spacing for eutectic solidification. These microstructural properties are empirically related to the solidification time and the rate of solidification respectively as [77, 78]

\[
\lambda_2 = 13.05 \times t_{SL}^{1/3} \quad (5.6)
\]

\[
\lambda_L^2 V_g \times 10^{-11} = 3.55 \times 10^5 \quad (5.7)
\]

where \(\lambda_2\) and \(\lambda_L\) are in µm representing the SDAS and lamellar spacing respectively, \(t_{SL}\) is the solidification time in s and \(V_g\) is the velocity of the solidification front in ms\(^{-1}\).
Figure 5-41 Eutectic Distribution within the Solidified Splat
Figure 5-42 show the dependence of these properties on Re and Ste. The SDAS is observed to decrease with increase in impact velocity which is a reflection of faster cooling caused by a wider contact area. Increase in Ste also results in a similar observation for the same reason. It is seen from Equation 5.7 that estimation of lamellar spacing requires information about propagation of the solidification front. Actual propagation speed is a microscopic quantity and its calculation is outside the scope of the present study. However, an approximate average speed can be determined using the following assumptions: the contour of $g = 0$ corresponds to the solidification front and the rate of eutectic growth is equal to the overall solidification rate. Macroscopic front velocity can easily be determined from the numerical results. Using the same value in Equation 5.7, the lamellar spacing is evaluated and is shown in Figure 5-42. (Note that the constant in Equation 5.6 is linearly interpolated from the values corresponding to Sn 5 wt% Pb and Sn 15 wt% Pb.). The average front velocity is inversely proportional to the solidification time and the corresponding effect on eutectic microstructure is a wider lamellar spacing.

Figure 5-42 SDAS and Eutectic Lamellar Spacing for different Impact Conditions
5.3.3 Effect of Subcooling

The Stefan approximation or Madejski’s model and its derivatives do not account for the effects of subcooling. As observed from previous results, substrate temperature may have significant effects on splat morphology and solidification behavior. A reference case with Ste = 0.667 and Re = 7000 is considered here to study the effects of subcooling by varying the substrate temperature. The minimum substrate temperature considered was 298 K while the upper bound of the range is limited by the eutectic temperature. The splat shapes observed are shown in Figure 5-43. Minor variations are observed due to slower cooling rates for high temperature substrates. The spreading of the spat is plotted versus dimensionless time in Figure 5-44 while the maximum spread factors at complete solidification are shown in Figure 5-45.

Figure 5-43 Splat Shapes for different Substrate Temperatures
The effect of lower subcooling can be visualized as an extended time scale for solidification while the spreading time scale remains unchanged.

Figure 5-44 Effect of Substrate Temperature on Spread Factor

Figure 5-45 Maximum Spread Factors for different Degrees of Subcooling

The effects of slower cooling rates can also be seen in the solidification and microstructural properties of the splat. The average eutectic fraction in the splat is shown.
in Figure 5-46. It is observed that the amount of eutectic in the droplet reduces drastically with subcooling. These results provide a way of enhancing the material properties of the solidified part by changing the operating conditions to meet the requirements. Moreover, the desired outcome would be achieved with minimum morphological alterations.

![Figure 5-46 Eutectic Fractions for different Degrees of Subcooling](image)

**5.3.4 Effect of Initial Concentration**

The use of a coupled temperature solute model facilitates the study of the effects of initial alloy composition on the overall splat formation. Initial concentrations of 10, 20 and 30 wt %Pb were considered for the present analysis. The splat shapes observed after complete solidification are shown in Figure 5-47. It is seen that the effects of alloy composition are more pronounced at low degrees of superheat and slower impacts. Under such circumstances, the overall solidification time is much smaller than at high Ste. The role of solidification in arresting the droplet deformation leads to profound differences in splat morphologies.
Solidification times for all cases are shown in Figure 5-48. Referring to Equation 2.25, it is clear that the mushy range diminishes as the composition approaches eutectic. The specific heat augmentation method briefly outlined in the previous section helps in understanding the solidification behavior at varying concentrations.
The liquidus temperature at $C_o = 10 \, \text{wt} \% \text{Pb}$ is 492.5 K while that at 30 \, \text{wt} \% \text{Pb} is 466.4 K. Correspondingly, the mushy range reduces from 36.5 K to 10.4 K and Equation 2.28 indicates that the effective specific heat during phase change, $C_{pm}$, increases approximately by a factor of 3.5 causing slower cooling when the initial concentration is increased. The analytical predictions made above are however, based on negligible superheat of the droplet. As the Ste is also increased, the spread factor and contact area become dominating factors and lead to marginally faster cooling even at higher concentrations.

The spread factors are shown in Figure 5-49 and it is observed that initial concentration has minimal effect on the splat morphologies. These findings are employed in the process of solder bumping using a eutectic solder. Slower cooling rate helps in the formation of eutectic microstructure having superior properties pertaining to electrical interconnects. The progressively increasing eutectic fractions in the droplet are shown in Figure 5-50 for varying impact conditions. Moreover, the liquidus temperature is the lowest at the eutectic point making the alloy easier to melt and reducing the degree of superheat required at impact.

![Figure 5-49 Spread Factors for varying Alloy Compositions and Impact Conditions](image-url)
5.3.5 Effect of Contact Resistance

As mentioned and shown earlier in section 4.1, contact resistance plays an important role in determining the splat morphology as well as solidification characteristics. Attinger et al. [9] and Wang et al. [66] emphasize the influence the contact resistance has on the overall process. A parametric study with different values of heat transfer coefficient for the case of a Sn-Pb alloy droplet deposition has been performed. As seen earlier, imperfect contact between the liquid alloy and the substrate surface results in elevated interface temperature and subsequently to slow cooling. Figure 5-51 shows the difference between interface temperatures and the isothermal substrate temperature for various heat transfer coefficients. If the contact resistance is high, it is observed that the interface temperatures are almost 100 to 200 K higher than the substrate temperature. The onset
of solidification is thus delayed substantially or theoretically, if the resistance is infinite, the substrate surface may even behave as an adiabatic boundary condition. The fluctuation in the interface temperature at approximately 1.5 ms in Figure 5-51 is because of the droplet remains in liquid state throughout the impact and recoil stages. The droplet thickness at the instant of maximum spread is very small and results in drastically large temperature gradient at the center line. The result is a very fast cooling and a sudden drop in temperature which is evident from the following figure. For lower contact resistances, such a situation is not encountered as the solidification progresses comparatively fast and is initiated during impact itself so that the droplet recoil is arrested.

As a consequence of the delayed solidification process, the droplet spreads out farther on the substrate surface and yields a higher spread factor. Figure 5-52 and Figure 5-53 depict the influence of contact resistance on the splat morphology. The splat thickness and maximum spread are greatly influenced by the effective heat transfer coefficient. The effects are more pronounced if the impact velocity is high and a larger
contact area is available for the heat transfer. A bump formation shows little but insignificant differences. Moreover, it is also observed that the influences of heat transfer coefficient on the splat morphology become less severe as the contact resistance, indicating the approach to a perfectly isothermal surface.

Figure 5-52 Effect of Contact Resistance on the Solidified Splat Shapes

Figure 5-53 Effect of Contact Resistance on Spread Factor
Chapter 6
Conclusion

The process of droplet deposition has been studied with a homogeneous mushy region model and a coupled temperature solute model. The multiphase fluid flow, heat transfer and solute transport equations are formulated using a finite volume approach and solved in a continuum domain. The factors governing phase transformation are closely examined. Information from the phase diagrams is utilized in a generalized non-equilibrium solidification modeling method. Microscopic phenomenon such as back diffusion and species segregation are also accounted for. The overall scheme was validated through experimental findings regarding droplet morphology and solidification behavior and an excellent agreement was found.

A homogeneous mushy region assumption is mathematically formulated to give a precise macroscopic description of the droplet deposition process. The key aspect of this formulation is its applicability to a wide range of materials ranging from pure metals to complex multicomponent alloys and the capability to predict substrate remelting. The scheme was applied to study the deposition of an SS304 droplet on a similar substrate under varying impact conditions. The splat morphology and cooling behavior are accurately modeled. Moreover, the consideration of the actual convection terms in the energy equation prove significant in predicting the overall solidification characteristics as well as local microstructural properties. A comparison with an augmented diffusion limit model [22] was made to evaluate the role of convection in the energy equation. The observations emphasize the convection dominant nature of the problem. A good agreement is obtained between the experimental findings and the results from the present study in terms of solidification characteristics as well as the thermal behavior and
microstructural properties predicted empirically. The augmented diffusion limit comes with serious limitations while predicting the overall time scale of the problem and the thermal profiles within the splat. The consequences of neglecting the impact dynamics as assumed in some of the previous works [22] are evaluated quantitatively which reveals that assumption of instantaneous spread is only valid under certain restrictive circumstances. The findings from the present analysis indicate that spreading time scale is approximately 30% of the overall solidification period over a range of parameters. Decoupling the spreading and solidification behaviors may not yield accurate results. A parametric study relating the effects of impact velocity and substrate preheating provide means of optimizing the process. Varying remelting depths and microstructural properties are observed by controlling the spread factor of the plat through these parameters.

Although a computationally efficient means of predicting the phase change characteristics, the homogeneous mushy region model fails to address the vital factors governing phase transformations in case of solid solutions or alloys. Alloy solidification is typically characterized by equilibrium phase diagrams. The homogeneous mushy region model makes use of the phase diagram in determining the solidus and liquidus temperatures without accounting for the instantaneous variations in species composition arising throughout the process. The limitations are evident when applied to complex alloy systems such as eutectics. Moreover, the fundamental assumption of a linear relation between latent and sensible heats eliminates the role of solute segregation in the governing equations. Subsequently, the peculiar eutectic behavior remains hidden and its microstructural analysis is not possible. To overcome the limitations, a coupled temperature-solute governing relationship is formulated and solved numerically. Various non-equilibrium solidification theories are built into the model through an approximate back diffusion parameter. Performance of these theories is briefly evaluated, although the
process of droplet deposition is best modeled through a rapid solidification assumption owing to the extremely small time scale for solidification.

The splat morphology, solidification characteristics and microstructural properties of a Sn-Pb alloy droplet being deposited on an isothermal substrate are analyzed under varying impact conditions. The splat morphology is highly dependent on the impact velocity and droplet temperatures. The formation of a solder bump or a splat is characterized mainly by the impact Reynolds number while the Stefan number has little contribution in this respect. A study of droplet oscillations reveals that solidification process plays a decisive role in determining the splat shape by arresting the recoil and oscillatory behavior. The extent of droplet spreading or the droplet-substrate contact area is observed to have the most governing influence on the solidification characteristics. A higher impact velocity and droplet temperature results in a greater spread factor due to increased initial kinetic and thermal inertia. However, splashing of the droplet is observed in extreme cases which limit the scope of the present analysis to \( \text{Re} < 14000 \) and \( \text{Ste} < 10 \). A brief comparison with the approximate analytical solutions proposed in the literature exhibits their limitations. The negligence of impact dynamics and viscous losses restrict the application of such analytical solutions to high Reynolds number only.

The effects of concentration profiles in determining the solidification characteristics are also examined. The radial convection fluxes are found to have a stronger influence on the concentration profiles. The classical results are retained under zero velocity environments present at the stagnation point. The results of liquid concentration and temperature values when transformed onto a phase diagram depict the implicitly followed local equilibrium condition. A comparison with the homogeneous mushy region approximation makes the fundamental difference more evident. One of the major strengths of this model is the capability to predict eutectic formations within the
splat and studying the complete solidification behavior through a special treatment. In the present work, the eutectic fraction is identified from the coupled governing equations following which, a homogeneous mushy region treatment is applied to the eutectic portion. The treatment makes it possible to simulate the complete phase change behavior rather than having to terminate the model at the eutectic point or extrapolate the coupled equations which violate the physics of eutectic formation. The microstructural properties such as lamellar spacing of eutectic and secondary dendritic arm spacing of the columnar growth are empirically related to the solidification characteristics. Parametric studies reveal that desired solidification properties could be achieved with minimal alterations in splat morphology by regulating the extent of subcooling. Initial alloy composition certainly affects the overall process as it is directly related to the mushy region and equilibrium temperature through the phase diagram. Careful consideration of contact resistance is needed as it plays a vital role irrespective of the impact conditions.
Chapter 7
Future Work

7.1 Model Improvements

Free Surface Convection

As mentioned in the previous sections, an adiabatic free surface boundary condition is used for the present analysis. The convection heat losses to ambient are approximately 1% of the total heat content of the droplet and prove insignificant. However, modern day research has lead scientists to develop sophisticated small scale fabrication techniques such as 3D metal printing [79]. To stabilize the liquid jet or a sequence of droplets, the process is carried out in a liquid environment to enhance the cooling rates. Convection is the primary model of heat loss in such applications and most of the temperature drop is observed even before impact. Furthermore, inflight heat transfer and cooling analysis of droplets or jets also require free surface convection.

The method of implementing free surface convection is rather simple given the implementation of a PLIC free surface construction. Heat losses due to convection can be accounted for through an extra source term in the energy equation given by

$$S_{\text{conv}}^o = -2\pi r_p h_{\text{conv}} L_{\text{surf}} (T_p^o - T_a)$$

(7.1)

where $h_{\text{conv}}$ is the convection heat transfer coefficient determined from the Nusselt number, $T_a$ is the constant ambient temperature and $L_{\text{surf}}$ is the interface length within a computational cell. The equation needs to be modified to make is compatible with the enthalpy formulation. Determination of $L_{\text{surf}}$ becomes the objective here. Suggested equations for the interface lengths corresponding to the configurations given in Figure 3-2. are as follows.
\[ L_{\text{surf}} = \begin{cases} \sqrt{(a_E + a_W)^2(\Delta y)^2 + (a_N + a_S)^2(\Delta x)^2} & \text{for } \sum_{1}^{4}[aw_i] = 2 \\ \sqrt{(a_E - a_W)^2(\Delta y)^2 + (a_N - a_S)^2(\Delta x)^2} & \text{for } \sum_{1}^{4}[aw_i] = 3 \\ \sqrt{(1 - a_E)^2(\Delta y)^2 + (1 - a_W)^2(\Delta y)^2 + (1 - a_N)^2(\Delta x)^2 + (1 - a_S)^2(\Delta x)^2} & \text{for } \sum_{1}^{4}[aw_i] = 4 \end{cases} \] (7.2)

where \( \sum_i[aw_i] \) denotes the number of cell faces wetted (completely or partially) by the fluid.

**Dynamic Contact Line and Contact Angle**

The wetting characteristics during solidification of a molten metal droplet has been studied by some researchers [68] experimentally and it proves to be a wide area of research within itself. Imaging techniques are used to capture the dynamic contact angles during spreading and recoiling and the only outcome is an empirical description which is case specific. The advancing solidification front and a drastically deforming droplet make the measurement and formulation of contact angle and wetting line challenging. In the present study, the contact angle is always computed at the bottom wall of the domain with the assumption that the dynamic spreading behavior of the splat is primarily governed by solidification and that the dynamic contact angle is neglected. At low Re the spreading and recoil effects are clearly visible. However, the difference between the advancing and receding contact angles is quite insignificant. However, Anderson et al. [80] outline a simple procedure to base the contact angle measurement on the dynamic contact line or the solidification front. Their calculations assumed a simplified horizontal
solidification front which is physically improbable. However, a similar idea can be implemented to accurately measure the contact angle at the dynamic contact line.

Figure 7-1 Dynamic Contact Line and Contact Angle

Figure 7-1 shows a situation where the solidification front appears within the splat. In the simplified model on the right [80], the solidification front is always assumed horizontal and the liquid – vapor and liquid – solid angles i.e. $\varphi_l$ and $\varphi_s$ respectively are easily measured using the bottom domain wall as the reference. However, the measurement of these quantities is much more difficult if the curvature of the solidification front is accounted for. In such situations, the actual angle definition requires information about the orientation of the solidification front or the dynamic contact line given as

$$\vec{n}_s = -\frac{\nabla \theta}{|\nabla \theta|}$$  \hspace{1cm} (7.3)

where $\vec{n}_s$ is normal to the solidification front. Using these definitions and the dynamic growth angle assumption outlined by Anderson et al. [80], the effects of dynamic contact line can be incorporated. Moreover, Attinger et al. [68] also indicate the apparent contact angles observed through their experimental studies which can be integrated with the dynamic growth angle assumption to improve the model accuracy.
Better Eutectic Solidification Treatment

The approximate solidification treatment employed in the present work provided satisfactory results under varying impact conditions and alloy compositions. However, a set of test simulations were performed with different phase diagrams and a cause for minor inaccuracies was identified. Figure 7-2 shows an artificial phase diagram with varying liquidus slopes. The partitioning coefficients are chosen such that the solubility limit for the α phase and the ratio $C_\alpha/C_{eutt}$ remains unchanged. Note that the maximum value of eutectic concentration possible is 100 wt % while the minimum is $C_\alpha$. The present model assumes the same fixed artificial mushy region for all the three cases shown in Figure 7-2. However, the eutectic fractions obtained in each case will be different, with the maximum in case C. Therefore, the solidification rate for case C will be the slowest. To have a truly homogeneous representation for a eutectic solidification, it is more appropriate to have the artificial mushy range given by

$$\frac{\delta_A}{\theta_{euttA}} = \frac{\delta_B}{\theta_{euttB}} = \frac{\delta_C}{\theta_{euttC}} = \varepsilon$$  \hspace{1cm} \text{(7.4)}$$

rather than $\delta_A = \delta_B = \delta_C = \varepsilon$.

Figure 7-2 Artificial Phase Diagram with Varying Partitioning Coefficients
Modeling Phase Change in Multi-Component Alloys

Although binary eutectic alloys are most commonly used in applications, there are certain processes which require multi-component alloy systems. Addition of metals in alloys is done to achieve desired material properties and microstructure. In such applications solidification is governed by segregation of various species and the modeling of species components becomes difficult. The works of Boettinger et al. [81] do address this aspect along with the multiphase reaction and thermodynamic equilibrium paths. Other noteworthy contributions come from [30, 82, 83, 84, 85] with attempts to model segregation of species under equilibrium solidification. However, rapid solidification modeling of multicomponent alloys is still a potential research area.

7.2 Applications

Liquid Phase 3D Printing

Wang and Liu [73] and Zuo et al. [79] proposed a highly robust and considerably versatile 3D printing technology in liquid environment. Metals and alloys have very high specific heats and relatively low molten state viscosity in comparison with the polymers used in conventional 3D printing technologies. This makes the problem of stability of the jet or droplet more crucial in applications requiring metal printed parts. An alternative according to [73] was to carry out the process submerged in a liquid such as water or ethanol. Certain important properties of water and ethanol relative to air are given in Table 7-1 [73].

It is evident from the thermal properties that the rate of cooling is substantially high in a liquid environment. Moreover, a high specific heat capacity makes it easier to maintain an isothermal ambient condition. Such a technique makes the solidification of the jet or droplet almost instantaneous without extensive deformations.
Table 7-1
Liquid Phase Properties Relative to Air

<table>
<thead>
<tr>
<th></th>
<th>Water</th>
<th>Ethanol</th>
</tr>
</thead>
<tbody>
<tr>
<td>Thermal Conductivity ($k/k_{air}$)</td>
<td>23.05</td>
<td>9.27</td>
</tr>
<tr>
<td>Viscosity ($\eta/\eta_{air}$)</td>
<td>55.87</td>
<td>67.04</td>
</tr>
<tr>
<td>Density ($\rho/\rho_{air}$)</td>
<td>828</td>
<td>655</td>
</tr>
<tr>
<td>Heat Capacity ($c/c_{air}$)</td>
<td>4.16</td>
<td>2.41</td>
</tr>
</tbody>
</table>

A critical numerical challenge however is to account for in-flight resistance and heat loss by the liquid metal or alloy droplet. Superior thermal properties of liquid environment come with the penalty of flow resistance. The model improvement suggested in section 7.1.1 is needed to model such behavior. Moreover, additional body force terms also need to be introduced in the flow governing equations.

**Spin Coating with Metals and Alloys**

Spin coating is a technique used for fine coating of polymer substance on a substrate to act as photoresist. A similar approach has also recently been adopted to acquire fine metal coatings on substrates to improve surface and electrical properties of small devices. A molten metal or alloy droplet is made to impinge on a rotating substrate. The centrifugal forces help in achieving extremely high spread factors of up to 4 with film thickness of the order of microns in a short period of time. Figure 7-3 shows the effects of angular speed of a rotating disk on the splat morphology.

Although the axisymmetric property of droplet deposition is not truly retained in the process, approximate treatment of the radial body force representing the spin effect
can be introduced in the Navier Stokes equation. The steady state spin of the substrate
can easily be accounted for while the spin up process needs further consideration. Spin
deposition process is a classic example of the simultaneous solidification and spreading
behavior of molten metal droplets.

![Figure 7-3 Evolution of Splat Shapes on a Rotating Disk](image)

**Lead Free Solder**

Modern day electronics has almost completely adapted to a lead free
environment. Newer solder alloys such as Sn-Bi, Sn-Ag-In, Sn-Ag-Bi etc. are replacing
the conventional Sn-Pb alloy while the search for a perfect alloy composition still
continues. Material properties such as strength, ductility and electrical conductivity are
greatly influenced by the microstructural developments throughout the deposition
process. Solidification characteristics of the new solder alloys are not yet well established
even through simplified theoretical models. The non-equilibrium solidification modeling of
these elements will provide great means of optimizing the process as well as the alloy
composition for a wide range of applications involving solder bumping for electrical
interconnects.
Appendix A

Grid Refinement for SS304/SS304 Deposition
Grid refinement tests for SS304 impact on SS304 substrate are shown in the section. A sample coarse grid structure is given in the following figure. The highest grid density is centered at the droplet substrate surface to accurately capture substrate remelting and impact dynamics. Grid tests were performed with 50% reductions in subsequent cases and the results of the free surface, maximum droplet temperature and substrate remelting were analyzed and are presented in the current section.

A Quadratically Increasing Grid Structure
Grid Convergence for SS304/SS304 Cases
Appendix B

Grid Refinement for Sn-Pb Deposition
The grid converge tests for SnPb alloy cases are given in the following figure. A uniform square mesh has been used for the studies.
Appendix C

Program Flow Charts
Numerical Algorithm for Enthalpy Solution
CLSVOF Algorithm [55]
Appendix D

Program Execution
The current section outlines the various aspects of numerical program executing the model. The program is written in FORTRAN and executed on a High Performance Computing platform operating on UNIX. The High Performance Computing facility at the University of Texas at Arlington and the Texas Advanced Computing Center at The University of Texas at Austin were used for the simulations. Following are the four files required for the execution with their corresponding tasks.

<table>
<thead>
<tr>
<th>File</th>
<th>Task</th>
</tr>
</thead>
<tbody>
<tr>
<td>bjob</td>
<td>A set of instructions for the Load Sharing Facility (LSF) on HPC platform. It is a batch script written to upload a case.</td>
</tr>
<tr>
<td>input</td>
<td>Contains a set of parameters describing a particular case.</td>
</tr>
<tr>
<td>filenam.dat</td>
<td>Contains the total number of output files to be generated and the file name of each.</td>
</tr>
<tr>
<td>tcripple.exe</td>
<td>The executable file generated after compiling the program. It contains flow information for the numerical calculations and all the subroutines required. ripple.exe was the basic multiphase flow solver while tcripple.exe includes the phase change module and enhanced interface tracking and construction schemes.</td>
</tr>
<tr>
<td>or ripple.exe</td>
<td></td>
</tr>
</tbody>
</table>

The collection of various subroutines written to perform specific tasks is logically integrated in the main program which upon successful compilation produces the executable called ripple.exe (only flow solver) or tcripple.exe (flow solver coupled with phase transformation). The bjob file is used to submit a case to be run on HPC. The parameters and data required for the numerical experiment are passed from the input file. A set of output files are generated as per the instructions provided in filenam.dat. Sample input and output files are shown below with a brief discussion.
Sample Input File:

Droplet_impact (SnPb Test) [mm,ms,mg,K]

$numparam
  alpha=1.0,
  autot=1.0,
  conserve=.false.,
  delt=1.0e-5,
  dtmax=1.0e-3,
  twfin=200000.0,
  con = 0.3
  fculim=0.5,
  idiv=1,
  dmpdt=3000000.0,
  prtdt=1000000.0,
  pltdt=0.01,
  sym=.true.,
  kt=1,
  kb=2,
  kl=1,
  kr=3,
$end

$fldparam
  gy=-9.81e-3,
  icyl=1,
  isurf10=1,
  psat = 0.0,
  xnu=3.14288e-4,
  rhof=7.0,
  sigma=0.566,
  vi=1.0,
  cangleb=135.0,
$end

$mesh
  nkx=1,
  xl=0.0,2.2,
  xc=1.1,
  nxl=55,
  nxr=55,
  dxmn=0.02,
  nky=1,
  yl=0.0,1.28,
  yc=0.64,
  nyl=32,
  nyr=32,
  dymn=0.02,
$end

$obstcl
  nobs=0,
$end

$freesurf
  nfrsurf=2,lequib=0,
  fcl(1)=-1.0, ifh(1)=1,
  fa2(2)=1.0, fb1(2)=-1.26, fb2(2)=1.0, fc1(2)=0.0944, ifh(2)=0,
$end

$graphics
  plots=.true., dump=.false.,
  iout = 0,
  iysymplt=1,
$end

$heateq
  heat = .true.,
  ischeme = 3,
  tid = 604.5,
The first line contains the problem name, following which are nine name lists. Name list is a group of variables containing a specific type of information as shown below.

**Numparam:** Numerical parameters. Variables defining initial conditions, boundary conditions, initial time steps, plotting frequency etc.

**Fldparam:** Fluid parameters. Variables defining properties such as density, viscosity, surface tension, contact angle, impact velocity etc.

**Mesh:** Mesh parameters. Variables defining mesh properties such as convergence point, minimum and maximum domain sizes and sub-meshes for non-uniform grids.

**Obstcl:** Obstacle parameters. Coefficients defining the conic function representing the surface of obstacles within the domain.

**Freesurf:** Initial free surface parameters. Coefficients defining the conic function representing the fluid free surface within the domain at \( t = 0 \).

**Graphics:** Originally contained plotting instructions. Currently, post processing is
performed using MATLAB. Variables in this list are thus, available for custom user defined operations.

Heateq: Energy Equation Parameters. Variables defining thermal properties such as conductivity, specific heat, mushy range, initial temperatures etc.

Coupled: Logical variables for selecting the interface tracking scheme.

Soleq: Solute Transport Parameters. Variables defining alloy phase diagram properties, initial composition and back diffusion model.

Sample Output File:

9.70340E-001 ← Time of plot
2,110 ← First and Last Real Cell # in x or r direction
2, 64 ← First and Last Real Cell # in y or z direction

0.00000E+000
2.00000E-002
4.00000E-002
6.00000E-002
Horizontal or Radial Locations of Cell Faces

0.00000E+000
2.00000E-002
4.00000E-002
6.00000E-002
Vertical or Axial Locations of Cell Faces

0.00000E+000,-5.01173E-006, 1.00000E+000, 1.00000E+000, 4.49940E+002, 3.81000E+001
-3.55794E-005,-3.34617E-004, 1.00000E+000, 1.00000E+000, 4.99400E+002, 3.81000E+001
-4.49273E-003, 4.52808E-003, 1.00000E+000, 9.41416E-001, 4.60416E+002, 3.47399E+001
-1.69689E-002, 5.77387E-003, 1.00000E+000, 8.22968E-001, 4.71851E+002, 2.58693E+001
8.18075E-003,-1.34855E-002, 1.00000E+000, 5.56255E-001, 4.85013E+002, 1.56872E+001
8.80911E-002,-3.41249E-003, 1.00000E+000, 2.50708E-002, 4.92241E+002, 1.00000E+001
9.93670E-002,-1.79820E-002, 1.00000E+000, 0.00000E+000, 4.99253E+002, 1.00000E+001

U or u, V or u, F, g, T, C
Velocity Components, VOF Function, Solid Fraction, Temperature, Concentration
References


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Biographical Information

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