NUMERICAL SOLUTION TO PHASE CHANGE PROBLEM: APPLICATION TO SUSPENSION PLASMA SPRAY COATINGS

by

Amirsaman Farrokhpanah

A thesis submitted in conformity with the requirements for the degree of Doctor of Philosophy
Graduate Department of Mechanical and Industrial Engineering
University of Toronto

© Copyright 2017 by Amirsaman Farrokhpanah
Abstract

Numerical Solution to Phase Change Problem: Application to Suspension Plasma Spray Coatings

Amirsaman Farrokhanah
Doctor of Philosophy
Graduate Department of Mechanical and Industrial Engineering
University of Toronto
2017

A study of flight, impact, and solidification of molten ceramic droplets generated by suspension plasma spraying (SPS) is conducted. A three-dimensional model is developed for predicting behavior of droplets generated by SPS impacting a solid substrate. The model combines Smoothed Particle Hydrodynamics (SPH) integral interpolations with enthalpy formulations to capture the phase change process (solidification/melting). Surface tension is modeled as an internal force between all particles, that will be canceled in the bulk of fluid and generate unbalanced surface tension forces near the free surface. A Finite-Volume solver is also used to predict properties of the droplets at the time of reaching the substrate, i.e. temperature, velocity, and diameter. This solver uses discrete phase models to track flight, evaporation, and atomization of suspension droplets injected into plasma flow. Effects of high temperature gradients and non-continuum on solid particles in plasma flow are taken into account.

Results are presented here in three steps. First, the newly developed numerical models for capturing solidification/melting in SPH are validated among various experimental, analytical, and numerical results from literature. These phase change models mainly fall into two main categories: (1) inclusion of latent heat as a source term
in the enthalpy equation, (2) inclusion of latent heat by modifying the effective heat capacity in the enthalpy equation. Results confirm accuracy and robustness of the new methods.

Secondly, results for droplet generation in SPS are presented. Effects of different parameters on droplets flight and impact are investigated. The goal here is to find suitable operating conditions for the plasma torch and injection process that guarantee impact of high quality droplets on the surface. Effects of injector parameters like injection location, flow rate, and angle along with effects of change in physical properties of droplets are studied.

Finally, the newly improved SPH model is used to predict impact of molten ceramic droplets that are collected on the substrate. These cases include predictions for the spread factor and droplet behavior based on their impact velocity and temperature. Results are used to explain different scenarios happening in substrate coating using SPS.
To my lovely wife

Reihaneh
Acknowledgements

I would like to express my sincere gratitude to my supervisor, Professor Javad Mostaghimi, for his continuous encouragement and nonstop support, and letting me to explore my scientific curiosity.

I would also like to thank Professor Markus Bussmann for his help and generous guidance. My sincere thanks also go to Professor Sanjeev Chandra, Professor Thomas W. Coyle, and Dr. Tony Murphy for their insightful comments.

Special thanks also to all my friends at Center for Advanced Coating Technologies (CACT).

This work was made possible through the financial assistance of the Government of Ontario through Ontario Graduate Scholarship (OGS) and the Glynn Williams Fellowship.
Contents

1 Introduction .................................................. 1
   1.1 Overview .................................................. 1
   1.2 Smoothed Particle Hydrodynamics (SPH) ............... 3
   1.3 Phase change: Solidification & Melting ............... 5
   1.4 Suspension Plasma Spraying (SPS) ..................... 7
   1.5 This Thesis .............................................. 10

2 Mathematical Formulation and Numerical Model ............. 14
   2.1 SPH Principles ........................................ 14
   2.2 Navier-Stokes equations .............................. 16
   2.3 Particle Tracking ...................................... 17
   2.4 Boundaries ............................................. 19
   2.5 Tensile Instability ................................... 21
   2.6 Time Marching .......................................... 22
   2.7 Latent heat evaluation ................................. 23
      2.7.1 Enthalpy Formulation with phase-change ........... 23
             Energy equation .................................. 23
             Temperature Calculation ......................... 24
             Kirchhoff temperature ............................ 25
3.3.1 Two-Dimensional Problem: Phase Change in the Corner of a Square .......... 61
      Comparison to Numerical Results ......................................... 63
      Effect of particle resolution .............................................. 63
      Effect of $\Delta T$ ............................................................ 64
      Effect of Smoothing Kernel .................................................. 66

3.3.2 Three-Dimensional Problems .............................................. 67
      Melting of a sphere .......................................................... 67
      Melting of a cone ............................................................. 68

3.3.3 Computational Cost ......................................................... 68

3.4 Choice of the Right Model ................................................... 74

4 Suspension Plasma Spraying (SPS) ............................................. 76
   4.1 Problem Specification ..................................................... 76
   4.2 Numerical Methodology .................................................... 77
      4.2.1 Torch Model .............................................................. 79
      4.2.2 Suspension Particle Model ............................................ 81
      4.2.3 Momentum ............................................................... 81
      4.2.4 Breakup model ......................................................... 84
      4.2.5 Heat transfer ........................................................... 85
      4.2.6 Evaporation of the liquid phase .................................... 86
      4.2.7 Melting of the solid content ....................................... 87
      4.2.8 Effective viscosity .................................................... 89
      4.2.9 Maximum packing fraction ($\phi_m$) .................................. 93
      4.2.10 $\phi_m$ calculation from empirical data ............................ 93
   4.3 SPS Particle Tracking ........................................................ 96
4.3.1 Torch Validation ........................................... 96
4.3.2 Effect of viscosity on breakup .............................. 97
4.3.3 Effect of viscosity model ................................. 98
4.3.4 Effects of Injection Parameters ............................ 100
4.3.5 Effect of Torch Operating Conditions ....................... 104
4.3.6 In-flight conditions ...................................... 106
4.3.7 Substrate Collections ................................... 114

5 SPS Droplet Impact & Solidification 122
  5.1 Problem Specification ........................................ 122
  5.2 Multiple Splat Overlaps ..................................... 125
  5.3 Spread Factor ............................................... 125
  5.4 Solidification Time and Heat Flux ............................ 136
  5.5 Predictions for Finished Coating ......................... 143
  5.6 Pore Formation ............................................ 144
  5.7 Conclusion .................................................. 167

6 Closure 170
  6.1 Conclusions ............................................... 170
  6.2 Recommendations for future Work ....................... 173

Bibliography 178
List of Tables

3.1 Smoothing kernels used in Enthalpy Formulation test cases . . . . . . . 55
3.2 Smoothing kernels . . . . . . . . . . . . . . . . . . . . . . . . . . . . 67

4.1 Summary of operating conditions . . . . . . . . . . . . . . . . . . . . 78
4.2 Materials properties . . . . . . . . . . . . . . . . . . . . . . . . . . . . 78
4.3 Torch conditions . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . . 97
4.4 Test case conditions . . . . . . . . . . . . . . . . . . . . . . . . . . . . 105
4.5 Effect of torch parameters . . . . . . . . . . . . . . . . . . . . . . . . . 106

5.1 YSZ particles’ properties at impact . . . . . . . . . . . . . . . . . . . 123
5.2 Average impact velocity ($\bar{w}$) and distance from substrate center ($\bar{r}$) for
different diameter ranges in case E (table 4.5). . . . . . . . . . . . 133
5.3 An estimate of the number of splat overlaps after impact on the substrate.137
List of Figures

1.1 Suspension Plasma Spraying (SPS) process. .................................. 3
1.2 SPH simulation of a single-phase drop using microscopic surface tension
forces (left), and a rendering of the reconstructed droplet surface (right). 6

2.1 Enthalpy variation for a material with a single phase change tempera-
ture (left) and phase change over a temperature interval (right). .... 24
2.2 Apparent heat capacity with a step jump for the latent heat. ....... 34
2.3 The smoothing function W. .................................................... 36

3.1 Formation of a 3D drop under surface tension from initial cubic shape 42
3.2 Calculated pressure jump of the drop at equilibrium ............... 43
3.3 surface tension coefficient (σ) for drops with different diameters. ... 43
3.4 Calculated pressure jump of drops at equilibrium for different mesh
resolutions. ............................................................................. 44
3.5 Calculated pressure jump of the drop at equilibrium. Particles are
initially arranged in the from of a droplet. PPD represents particles
per diameter. ............................................................................. 44
3.6 Water drops spreading on a surface with a constant contact angle.
Contact angles are 80° and 160°. ............................................. 45
3.7 Temperature profile across the slab with an initial unit temperature jump at $x = 0$. .......................................................... 47

3.8 Temperature profile across a slab with a temperature jump of 10 at $t = 0, 1ms, 7ms$, and $0.2s$. .............................................. 48

3.9 $L_2$ error values for temperature profile across a slab with a temperature jump of 10. .......................................................... 48

3.10 Temperature on the diagonal of the square for $t = 5, 25, 50, 75, 150$, and $250ms$ (from bottom to top, respectively). .................. 49

3.11 Difference in computed temperature values on the diagonal of the square plate, $E = \max (|T_{SPH} - T_{Fluent}|)$. .............................................. 49

3.12 Initial positioning of fluid/solid particles (hollow squares) and wall particles (filled squares). ............................................... 50

3.13 Problem description: fluid is assumed to be inside a Cuboid which is infinitely long in $z$ direction. At the start of simulation, temperature at walls is suddenly dropped to a value below freezing temperature. 51

3.14 Solidification front moving along the diagonal of a cross section of the cuboid for saturated liquid (compared against Cao et al. [52], Crowley [66], and Hsiao et al. [22]). ............................................... 52

3.15 Solidification front moving along the diagonal of a cross section of the cuboid for liquid above freezing temperature (Cao et al. [52], Hsiao et al. [22], and Keung [67]). ............................................... 53

3.16 Mesh resolution dependency. Figure shows solidification front moving along the diagonal of a cross section of a cuboid (which has a size of $2 \times 2$). Mesh resolution on the cross section has been changed from $10 \times 10$ to $100 \times 100$. .............................................. 54
3.17 First example repeated using different kernels from table 3.1. Figure shows solidification front moving along the diagonal of a cross section of the cuboid for saturated liquid. .......................... 55

3.18 Spread factor \((D/D_0)\) for a tin drop at 240°C with 2.7mm diameter impacting at 1m/s on a 25°C stainless steel substrate. .......................... 56

3.19 Divergence free initial velocity field for measuring effects of mushy region. 58

3.20 Particle positions in a \(1 \times 1\) rectangle at \(t = 0.16\) (s) colored by their liquid fractions. Mushy zone effected included using equation 2.77 (left) and equation 2.78 (right). .......................... 59

3.21 Solidification front (liquid fraction = 0.5) for equations 2.77 and 2.78 at \(t = 0.16\). ................................................................. 60

3.22 Problem description: fluid is assumed to be inside a Cuboid which is infinitely long in \(z\) direction. At the start of simulation, temperature at walls is suddenly dropped to a value below freezing temperature. 61

3.23 Initial particle positioning. Hollow circles show fluid particle positions, while filled circles indicate wall particles. ................................. 63

3.24 Solidification front position versus the analytical solution, for solidification in a corner. ............................................................ 64

3.25 Position of the solidification front on the diagonal cross section of the cuboid (resolution: \(100 \times 100\)) ............................................. 65

3.26 Position of the melting front moving along the diagonal of a cuboid \((30 \times 30)\) ................................................................. 66

3.27 Melting of a cuboid at 0.1, 0.4, 0.8, 1.2, and 1.44 seconds. Solid lines are the analytical solution. Solution resolution: 40 \(\times\) 40 (top) and 100 \(\times\) 100 (bottom). ................................. 69
3.28 Error of the solidification front position versus the analytical solution, for solidification in a corner, for various $\Delta T$. ......................... 70
3.29 $W(R, h)/h^3$ for the three smoothing kernels. ......................... 70
3.30 Error of the solidification front position versus the analytical solution, for three different kernels. ................................. 71
3.31 Melting of a sphere. Total time of melting versus Stefan number. Squares show results obtained using the model presented here, and the solid line is from the mathematical model of [77]. ............... 71
3.32 Melting of a sphere of radius 0.25, with a wall temperature $T_w = 1^\circ C$, at $t = 0, 0.0025, \text{and } 0.0065$ seconds. Left: un-melted particles; Right: temperature. ......................................................... 72
3.33 Melting of a cone. Interface position is plotted at different times: 0.05, 0.1, 0.25, and 0.5. Red crosses are the results of Ayasoufi et al. [78] obtained using a space-time conservation element and solution element (CE/SE) method. ................................. 73
3.34 Position of the solidification front on the diagonal cross section of the cuboid - comparing equations 2.59, 2.76, and numerical results of Cao [52]. ......................................................... 74
4.1 Schematic drawing showing torch and the particles that are being injected downstream. ......................................................... 77
4.2 Mushy zone effect on viscosity values of YSZ. ......................... 88
4.3 Effect of latent heat of melting on $C_p$ values. ......................... 89
4.4 Viscosity of YSZ-Water suspension under different concentrations of YSZ powder (from Arevalo-Quintero et al. [116]). Dashed line is the trend line. ................................. 92
4.5 \( 1/\mu_r \) for the YSZ-water suspension. Dashed line is the trend line. . . 94
4.6 \( (1 - \mu_r)^{-1/N} \) for the YSZ-water suspension. Dashed line is the trend line. 94
4.7 Effect of turbulence model on temperature profile, experimental results are from Brossa et al. [117] and numerical results from Bolot et al. [118] and Jabbari et al. [34]. .......................... 95
4.8 Effect of mesh resolution on temperature profile, experimental results are from Brossa et al. [117]. .......................... 95
4.9 Temperature profile for various torch conditions compared with experimental results of Brossa et al. [117]. .......................... 96
4.10 Velocity and temperature profile on the torch centerline against experimental and numerical results of Meillot et al. [119]. .......................... 98
4.11 Ohnesorge Number versus flight time for YSZ-water suspension particle injected into the domain. .......................... 99
4.12 Viscosity predictions for 8mol\% YSZ-water. .......................... 100
4.13 Particle standardized diameter and Reynolds number density distribution for different viscosity models. .......................... 101
4.14 Schematic drawing of the parameters defining injection conditions. . . 102
4.15 Particle counts on substrate for each test case. .......................... 104
4.16 Axial cross sections for the torch of case A. .......................... 107
4.17 Temperature profile of the plasma gas flow on the centerline of the torch (case E, Table 4.3). .......................... 108
4.18 Temperature profile for case A, plotted at cross sections perpendicular to the flow direction at distances of 3, 5, and 7mm from the nozzle exit (from top to bottom). .......................... 109
4.19 Snapshot of particle positions during flight for Cases A-E. Images on the right show particles during their flight from substrates point of view. Particles are colored based on their diameter (m).

4.20 Diameter, velocity, Reynolds number, Weber number, Ohnesorge number, and shear rates for particles during flight. Shown here for case A. The torch nozzle exit is located at X=0.02m.

4.21 Water mass fraction inside particles during flight for cases A-E.

4.22 Temperature of particles during flight for cases A and E.

4.23 Particle temperature distributions for test cases A-E upon impact on the substrate.

4.24 Particle diameter distributions for test cases A-E upon impact on the substrate.

4.25 Particles captured on the substrate 8cm downstream of torch for test case B collected over 3ms.

4.26 Particle normal and tangential velocity distributions for test cases A-E upon impact on the substrate.

5.1 Position and diameter of five YSZ drops impacting the substrate obtained from case E (table 4.5).

5.2 Solidification of droplet #1 on the substrate (table 5.1) colored by the height from the substrate.

5.3 Solidification of droplet #2 on the substrate (table 5.1) colored by the height from the substrate.

5.4 Solidification of droplet #2 on the substrate (table 5.1) colored by the height from the substrate.
5.5 Solidification of droplet #3 on the substrate (table 5.1) colored by the height from the substrate. ................................................................. 129
5.6 Solidification of droplet #4 on the substrate (table 5.1) colored by the height from the substrate. ................................................................. 130
5.7 Solidification of five drops on the substrate (table 5.1) colored by the height from the substrate. ................................................................. 131
5.8 Solidification of five drops on the substrate (table 5.1) colored by the height from the substrate. ................................................................. 132
5.9 Distribution of impact position classified based on particle diameter. . 134
5.10 Distribution of impact position classified based on particle diameter. . 135
5.11 Spread factor ($D/D_0$) for a YSZ droplet impacting the substrate at 50, 100, and 200 m/s ($t^* = tV/D_0$). .............................................................. 136
5.12 Impact and solidification time for a droplet at temperature of 3000 K with radius of 3 $\mu$m and velocity of 150 m/s. ................................. 138
5.13 Heat flux at the impact point of the droplet on the substrate as a function of non-dimensional time ($t^* = tV/D_0$), shown near the time of impact (figure on top) and afterwards (figure on bottom). ............. 140
5.14 Solidified splat from a droplet with 4.59 $\mu$m diameter, impacting the substrate at 175 ms with a temperature of 3517 K, shown at $t^* = 1500$. 141
5.15 Binary impact of two droplets with diameters of 4.59 and 6.8 $\mu$m at velocities of 175 and 231 m/s and temperature of 3517 and 3795 K. The position of impact is offset by 14 $\mu$m. ................................. 142
5.16 Finished surface of the substrate for cases A to E, colored by the height from the substrate’s surface. ......................................................... 145
5.17 Finished surface of the substrate for cases D and E taking particle temperatures into account, colored by the height from the substrate’s surface. 

5.18 Normal impact of droplet with a radius of 3µm on flat surface against surfaces with step sizes of 0, 0.12, 0.24, 0.36, and 0.54µm, from top to bottom respectively. Impact velocity is 150m/s and the initial temperature for the droplet and the substrate are 2985 and 300K. Current time for all cases is 0.1µs.

5.19 Normal impact of droplet with a radius of 3µm on flat surface against surfaces with step sizes of 0, 0.12, 0.24, 0.36, and 0.54µm, from top to bottom respectively. Impact velocity is 150m/s and the initial temperature for the droplet and the substrate are 3500 and 300K. Current time for all cases is 0.1µs.

5.20 Two-dimensional simulation of droplet with diameter of 18µm impacting a patterned surface at velocity of 250m/s.

5.21 Droplet with a diameter of 18µm impacting a patterned substrate at velocity of 250m/s and temperature of 3173K.

5.22 Closeup at \( t^* = 0.5 \) for a droplet with diameter of 18µm impacting a patterned substrate at velocity of 250m/s and temperature of 3173K.

5.23 Heat flux magnitude (W/m\(^2\)) for a droplet with diameter of 18µm spreading on a patterned substrate at velocity of 250m/s and temperature of 3173K at nearly \( t^* = 0.6 \).

5.24 Droplet with a diameter of 15µm impacting a patterned substrate at velocity of 250m/s and temperature of 3173K.

5.25 Droplet with a diameter of 15µm impacting a patterned substrate at velocity of 250m/s and temperature of 3173K.
5.26 Closeup of the solidification front movement inside the column voids, after impact of a droplet with diameter of 15µm on a patterned substrate at velocity of 250m/s and temperature of 3173K.

5.27 Void formation as a function of distance from impact point, against experimental and analytical results of Xue et al. [121].

5.28 Impact of seven droplets on a patterned surface at 150m/s, colored by temperature (K).

5.29 Coating process of a patterned substrate at \( t^* = 1.3 \), colored by temperature (K).

5.30 Coating process of a patterned substrate at \( t^* = 2.5 \), colored by temperature (K).

5.31 Coating process of a patterned substrate at \( t^* = 3.1 \), colored by temperature (K).

5.32 Coating process of a patterned substrate at \( t^* = 7.3 \), colored by temperature (K).

5.33 Coating process of a patterned substrate at \( t^* = 11.5 \), colored by temperature (K).

5.34 Heat flux magnitude (W/m²) between substrate and droplets at \( t^* = 1.9 \).

5.35 Magnitude of velocity (V) and acceleration components due to pressure (p), viscous (v), and surface tension (s) forces at \( t^* = 1.3 \).

5.36 Solidified material left in the voids at \( t^* = 15000 \).

5.37 Solidified material on the substrate with the assumption of having individual impacts and solidifications for each of the 7 droplets.

5.38 Estimation of substrate coverage using overlapping discs, shown for the case of five droplets in table 5.1.
5.39 Solidified material on the substrate for 1 million droplets obtained from numerical superposition of splats on the substrate (colored by height from the surface of the substrate). .................................................. 168

5.40 Growth of the area of the solidified material on the substrate as a function of time for 1 million droplets obtained from numerical superposition of splats on the substrate. .................................................. 169
Nomenclature

δ     Dirac delta function
ε     Emissivity
η     Thermal efficiency
γ     Heat capacity ratio
λ_{eff} Effective molecular mean free path
μ_r   Relative viscosity
Ω     Torch heat source volume
ϕ     Volume fraction of solid content in suspension droplets
σ     Stephan Boltzmann constant
C     Heat Capacity
C_D   Drag coefficient
C_p   Specific heat
D     Binary diffusion coefficient
d_p   Particle Diameter
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$F$</td>
<td>Thermophoretic force</td>
</tr>
<tr>
<td>$f_1$</td>
<td>Correction factor for strong temperature gradients</td>
</tr>
<tr>
<td>$f_2$</td>
<td>Correction factor for non-continuum effects</td>
</tr>
<tr>
<td>$F_D$</td>
<td>Drag force</td>
</tr>
<tr>
<td>$H$</td>
<td>Enthalpy</td>
</tr>
<tr>
<td>$h$</td>
<td>Convective heat transfer coefficient</td>
</tr>
<tr>
<td>$h_{fg}$</td>
<td>Latent heat of liquid phase</td>
</tr>
<tr>
<td>$i$</td>
<td>SPH target particle subscript</td>
</tr>
<tr>
<td>$j$</td>
<td>SPH neighbour particle subscript</td>
</tr>
<tr>
<td>$K$</td>
<td>Thermal conductivity</td>
</tr>
<tr>
<td>$K^*$</td>
<td>Ratio of fluid to particle thermal conductivities</td>
</tr>
<tr>
<td>$Kn$</td>
<td>Knudsen Number</td>
</tr>
<tr>
<td>$Kn^*$</td>
<td>Knudsen number based on effective molecular mean free path</td>
</tr>
<tr>
<td>$L$</td>
<td>Latent Heat</td>
</tr>
<tr>
<td>$M$</td>
<td>Molecular weight</td>
</tr>
<tr>
<td>$m_p$</td>
<td>Particle Mass</td>
</tr>
<tr>
<td>$Nu$</td>
<td>Nusselt Number</td>
</tr>
<tr>
<td>$Oh$</td>
<td>Ohnesorge number</td>
</tr>
<tr>
<td>$Pr$</td>
<td>Prandtl Number</td>
</tr>
</tbody>
</table>
$Pr_f$ Prandtl number evaluated at film temperature

$Q$ Torch volumetric heat source

$Re_d$ Reynolds number based on particle diameter

$Re_{DH}$ Reynolds number based on the hydraulic diameter

$s$ Direction of the temperature gradient

$Sc$ Schmidt Number

$T^*$ Kirchhoff Temperature

$t^*$ Non-dimensional time

$T_1$ Lower limit for melting temperature of an alloy

$T_2$ Upper limit for melting temperature of an alloy

$T_\infty$ Ambient temperature

$T_m$ Melting temperature

$T_p$ Particle Temperature

$T_{cell}$ Cell Temperature

$V_f$ Average thermal velocity

$W$ SPH Smoothing Kernel

$W^*$ Wet basis moisture content

$W^P$ SPH Position Smoothing Kernel

$W^T$ SPH Thermal Smoothing Kernel
We  Weber Number

$We_c$  Critical Weber Number

$\dot{\gamma}$  Shear rate

$[\mu]$  Intrinsic viscosity
Chapter 1

Introduction

1.1 Overview

Coating is the process of covering a surface with particular materials with the goal of protection or forming more advanced materials. Thermal spraying, one of the examples of the coating process, uses heat from combustion or electrical arc to melt solid materials. These molten droplets are then accelerated using gas jet to impact a destination substrate and solidify on its surface.

Thermal spray, originally driven by its aerospace applications, is now one of the most important methods for wear, thermal, and oxidation protection due to having a high deposition rate over a large area compared to many other coating methods [1]. Coating materials can be ceramics, alloys, polymers, and even composites which can be deposited over a wide range of materials such as plastic, steel, etc. Materials coated with these methods can possess many different properties such as high thermal resistance (thermal barrier coatings), wear protection by improving tribological properties of the surfaces that rub against each other, and antibacterial effects. Better understanding of this process will help in prediction of the properties and structure of the
An interesting type of thermal spray is the Suspension Plasma Spraying (SPS) process. SPS is emerging as a powerful coating technique for depositing high quality thermal barrier coatings. This process has been demonstrated in figure 1.1. Fine ceramic powders can be used to create stable suspensions in fluids like water or ethanol which are then injected into a plasma jet. The heat from plasma will evaporate the carrier fluid and eventually melt the solid content. The suspension droplets go through several breakups before evaporation is complete. The solid particles left after evaporation of the solvent can become aggregated before melting. The molten particles will then impact the substrate at high velocities.

Understanding effects of different parameters on the final quality of the droplets that impact the substrate helps in better prediction of the coating properties. Physical properties of the suspension, operating conditions of the plasma torch, along with injection parameters, i.e., position, angle, and speed, play important roles in this process. Interaction of the molten droplets upon impact and solidification on the substrate can also contribute to different depositions.

The goal here is to use numerical models to study the SPS process, from injection into the plasma flow till impact and solidification on the substrate. In this way, a numerical model is initially developed to study the trajectory of suspension droplets as they get injected into a plasma jet and experience evaporation of the solvent, variation of physical properties of the suspension, and melting of the solid ceramic particles. The predictions of this model is then used in a separate solver to study the impact and solidification process of the generated ceramic particles in more details. Combination of results are used for prediction of the final coated surface of the substrate.

The remainder of this chapter presents subjects related to this thesis: an introduction to Smoothed Particle Hydrodynamic method that includes review of available
surface tracking methods for multiphase studies, a review of numerical methods used in literature for capturing the phase change process, and finally, an overview of the Suspension Plasma Spraying process with literature review.

Figure 1.1: Suspension Plasma Spraying (SPS) process.

1.2 Smoothed Particle Hydrodynamics (SPH)

SPH is a Lagrangian mesh-free CFD method introduced in 1977 by Lucy [2] and Gingold and Monaghan [3], and was initially applied to astrophysics. Since 1977, many studies have been conducted on the accuracy and applicability of SPH to various
Chapter 1. Introduction

fluid problems [4]. The main advantage of SPH is its ability to handle complex geometries. The implementation of different boundary conditions in SPH is also straightforward. Moreover, SPH codes are easily parallelized.

Since 1977, SPH has been extensively used in simulating different physical phenomena in fields like astrophysics, fluid sciences, oceanography, ballistics, etc. One of the major subjects studied in SPH is interfacial flows. Practical studies such as tsunami simulations [5], simulation of floating bodies like ships [6], atomization of liquid jets [7], impact of liquid drops over surfaces [8], and other multiphase studies [9–11] are among them.

Many attempts have been made in simulating multi-phase fluid phenomena using SPH. Some of these multi-phase studies, like the work of Colagrossi et al. [12], have been performed without any specific treatments for modeling physical surface tension effects. These simulations produce acceptable results for interfacial flows. For droplet formation however, especially in micro-scale droplets, having a reliable surface tension treatment algorithm is necessary [12]. Hence, other multiphase studies have employed different methods for adding surface tension effects to multi-phase SPH like the methods proposed by Morris [13] and Hu et al. [9,14,15].

In SPH studies, surface tension forces are modeled in two different ways. In one, surface tension is included by means of macroscopic surface tension forces, like the methods proposed by Hu et al. [9] or Adami et al. [16], which are fundamentally based on the Continuum Surface Force (CSF) model of Brackbill et al. [17]. The main advantage of these models is the possibility of including surface tension coefficient as an input to the solution. These methods however are not applicable to single phase problems, as existence of at least two phases is needed for proper surface reconstruction. The other procedure for surface tension inclusion in SPH is by using microscopic methods, like the method proposed by Nugent et al. [18].
water droplet on a surface generated by this model has been shown in figure 1.2. In these methods, surface tension is added as an inter-particle force. These forces will cancel one another inside the bulk of fluid and will result in surface tension effects near the surface due to imbalanced cancellations. The advantages of these models is that they can be easily applied to both single and multi-phase cases. There is also no need of surface reconstruction in these methods as the surface tension forces are applied to all particles regardless of their position on the surface or bulk of the fluid. There is however a drawback of being mesh dependent [16], as the amount of this force needs to be adjusted based on how many particles are in equilibrium in each problem. There also exists no direct way of relating the magnitude of surface tension forces to the surface tension coefficient. These forces, hence, have to be calibrated for different problems based on the pressure jump they generate on the surface. From computational point of view, the fact that these microscopic forces need to be applied everywhere in the domain also makes the solution less efficient. However, microscopic methods can still be faster compared to CSF methods that need detailed surface reconstruction for calculation of the curvature on the surface. Also, the surface reconstruction procedures might impose time step constraints on the problem for ensuring a stable solution, where microscopic methods on the other hand could show a tendency to be more relaxed when it comes to the choice of time step. In this thesis, the microscopic procedures have been chosen for surface tension handling due to their efficiency in free surface single phase problems.

1.3 Phase change: Solidification & Melting

Heat transfer and phase change are of importance in many engineering applications, e.g. coating surfaces with paint or metal, freezing and thawing of food products, and
the casting of plastics and metals. There are a wide range of methods available for numerical prediction of transient phase change problems. Knowledge of the solid-liquid front position is of key importance. In many numerical simulations of phase change, the effect of latent heat is added as a source term to the heat transfer equation, like the models of Passandideh Fard [19] and Voller [20], or by modifying the heat capacity coefficient, as in Thomas et al. [21], Hsiao [22], and Dalhuijsen et al. [23]. The finite volume method was first used for the study of phase change heat transfer, followed by finite element methods. This transition was due to the flexibility of the finite element method for complex boundary conditions and implementations [23].

The use of SPH, or integral interpolations in a more general sense, has also been introduced for study of the phase change process.

Several studies have attempted to solve phase change problems using SPH. Cleary et al. [24,25] focused on explicit inclusion of latent heat after solving the enthalpy heat conduction equation. This method has been used in different cases such as casting simulations [26] and solidification of molten metal drops impacting a surface [27–29]. Monaghan et al. [30] modeled the solidification of pure and binary alloys, where phase
change was simulated by removing liquid particles and transferring their mass to a stationary grid of solid particles. The drawback of this method is the need for virtual particles, which results in a doubling of the number of particles inside the domain, and makes the method computationally expensive and difficult to implement into available solvers.

1.4 Suspension Plasma Spraying (SPS)

Suspension plasma spraying is emerging as a powerful coating technique for depositing high quality thermal barrier coatings. Fine ceramic powders can be used to create stable suspensions in fluids like water or ethanol which are then injected into a plasma jet. The heat from plasma will evaporate the carrier fluid and eventually melt the solid content. The suspension droplets go through several breakups before evaporation is complete. A detailed understanding of how process operating parameters affect the properties of the deposited coating is of great importance. Physical properties of the suspension, operating conditions of the plasma torch, along with injection parameters, i.e., position, angle, and speed, play important roles in this process.

Various experimental and numerical studies have been performed on suspension plasma spraying (SPS). The numerical and experimental study by Fazilleau et al. [31] focused on interaction between YSZ-ethanol suspension droplets and gas flow in DC plasma spraying. Suspension was injected at a 60° angle aiming the centre point of the nozzle exit. It was shown that plasma flow’s asymmetry due to liquid injection becomes deteriorated at 15mm downstream of the nozzle exit. Therefore, after 10 to 15mm, solvent in the suspension has completely evaporated and the solid contents are uniformly mixed with the plasma gas. Their study also pointed out that gas flow velocity variations induced by voltage fluctuations in 200µs time windows play important role
in stimulating droplet breakups. In the second part of their study [32], effect of gas fluctuations on solid particle flights was investigated using a simplified 2D model. Effects of plasma parameters on drag and heat transfer of solid particles was included using correction methods. Their model however assumed constant properties for injected particles. Obtained results suggested an optimum substrate standoff distance of $40 - 60 \text{mm}$. Waldbillig et al. [33] performed an experimental study on the effects of torch nozzle size, power, and plasma gas velocity on SPS YSZ coatings. They concluded that the lowest permeability in coatings can be achieved by using small nozzles and high plasma flow rates, while higher deposition efficiency is a result of lower flow rates with small nozzles. Jabbari et al. [34] performed a numerical study of Nickel-Ethanol suspension spraying. Similarly, Jadidi et al. [35] numerically focused on flight and impact of Nickel-Ethanol suspension droplets near the substrate and upon impact. Factors such as particle speeds and trajectories along with the shape and position of the substrate were studied. Their results showed that decreasing the substrate standoff location from 60 to $40 \text{mm}$ can increase particle temperature at substrate by 6%. Rampon et al. [36] studied application of SPS in producing solid-oxide fuel cells. Their experiment on spraying YSZ suspension in water reported a shift from mono modal particle size distribution to multimodal with the increase of gas Weber number from 5 to 24. More importantly, they concluded that the suspension breakups and droplet size distributions were affected by the viscosity of the suspensions. Their results showed that at high Weber numbers, Ohnesorge number, or viscosity of the suspensions, can become the dominant factor. It has to be noted though that their calculation of Weber and Ohnesorge numbers did not include change of physical properties during flight, i.e. viscosity of the suspension droplets was assumed to remain constant and equal to its initial value at injection time. Studies like this indicate that the physical properties of suspension materials can have
significant impact on the SPS process. For instance, they can tremendously control the atomization process. Suspension defragmentation is a result of domination of different forces acting on the droplets, i.e. (i) inertia and shear forces resulting from the relative velocities between the droplet and plasma gas, (ii) instabilities induced by surface tension forces resisting increase in the surface area of the droplets, and (iii) viscous forces that act by dissipating the instabilities. Three non-dimensional numbers can be used for evaluating the importance of each force compared to another, Weber number \( (We = \frac{\rho v_{rel}^2 d_p}{\sigma_p}) \), Reynolds number \( (Re = \frac{\rho v_{rel} d_p}{\mu_g}) \), and Ohnesorge number \( (Oh = \frac{\mu_p}{\sqrt{\rho_p \sigma_p d_p}}) \). \( \rho \) is the density, \( \mu \) is the dynamic viscosity, \( \sigma \) is the surface tension coefficient, \( d \) is droplet diameter, and \( v_{rel} \) is the relative velocity between a droplet and plasma gas. Subscripts \( p \) and \( g \) refer to properties of suspension droplets/particles and plasma gas, respectively. If any of the physical properties were to change, these non-dimensional numbers will be affected and this results have having different patterns of physical phenomena. Weber number plays an important role in determining the breakup regimes of suspension droplets. Its value can vary due to changes in gas density, relative velocity, droplet diameter, and surface tension. As the suspension jet is injected into the torch, the values for gas density and relative velocity will change. Meillot et al. [37] study shows that the jet’s Weber has a rapid growth upon entering the plasma flow due to high relative velocity and gas density. This value is then reduced with further penetration. As the jet reaches near the torch centre line, Weber will grow again slowly. Droplet diameter is typically controlled by problem parameters such as the injector’s size and geometry. Surface tension of droplets can also influence Weber values. For pure liquids, this value will remain constant as the droplets travel in the domain. For suspensions, however, surface tension values may differ. In the preparation process of the suspension, the addition of the solid content to the solvent typically results in a mixture with surface tension values
lower than the solvent’s original surface tension. It is however reasonable to assume that this lower surface tension value will not drastically change with the evaporation of the liquid content [36] and hence its effect on Weber number and breakup regimes is neglected in current scope. The viscosity of the suspension however, as already suggested by many researches [33, 38–41], goes through significant changes when the liquid content evaporates. The changing value of viscosity can potentially become a key factor in determining droplet breakup regime. While changes in viscosity do not affect Weber values directly, effect of viscosity on breakup according to experimental studies becomes important when $Oh > 0.1$ [42]. As the $Oh$ increases, the critical $We$ also increases which can be estimated for large $Oh$ as [43]

$$We_c = We_{c,Oh->0} (1 + 1.077Oh^{1.6})$$  \hspace{1cm} (1.1)

The critical $We$ marks the transition between breakup regimes [42] and theoretically drops will experience breakups for values larger than $We_c$. During the suspension flight, as the liquid content evaporates, the viscosity of the suspension increases. This increase of dynamic viscosity ($\mu_p$) will consequently increase Ohnesorge number ($Oh$). According to correlations like equation 1.1, this leads to increases of the $We_c$. In other words, as the droplets lose liquid solvent, they become more resistant to breakups.

### 1.5 This Thesis

This thesis presents study of flight, impact, and solidification of molten SPS droplets on a substrate. This process involves two main stages: (1) Suspension mixture is injected as a liquid jet into the plasma torch and is tracked while it goes through
evaporation, breakup, and melting before it generates molten particles that reach the substrate, (2) the particles that reach the substrate will impact and solidify on it. Combining these two stages into a single solver is numerically expensive as it demands resolving the domain in a multi-scale manner. Torch and its distance to the substrate are in orders of centimeters while the drops impacting the substrates are sub-micron. To capture the process completely, each step here has been simulated separately. Initially, the injection of suspension liquid in the torch has been modeled using the Finite Volume based ANSYS Fluent solver. Suspension liquid is tracked as it goes through evaporation and breakups. With the liquid content evaporated completely, the solid remains from the suspension are then tracked as they melt and form sub-micron particles that will reach the substrate at high speeds and temperatures. Results of these simulations are then fed into a Smoothed Particle Hydrodynamic (SPH) solver that covers the impact and solidification of the particles.

The objectives of this work include:

- implementation of a numerical solution that accurately calculates trajectory of suspension particles from injected port till reaching the substrate.

- development of a methodology for inclusion of effects of solvent evaporation on variations of thermal and physical properties, especially viscosity of the suspension.

- implementation of effects of plasma jet, such as Knudsen effect and high temperature gradients, on flights of solid particles traveling with the flow.

- a comprehensive study effect of different parameters, such as torch operating conditions and injection settings, on the trajectory and final quality of the molten ceramic droplets the reach the substrate.
Chapter 1. Introduction

- implementation of a robust and accurate model for tracking interface of sub-micron molten ceramic particles upon impact on solid substrate.

- development of numerical models for handling phase change problems by inclusion of latent heat released (absorbed) during solidification (melting).

- a comprehensive validation of the numerical model using available analytical, experimental, and numerical solutions in literature.

- application of the model to prediction of the final coating of the substrate over a brief spray time window.

The remainder of this thesis is structured as follows. Chapter 2 presents two different SPH formulations using integral interpolations are introduced for modeling the transient phase change problem. Chapter 3 presents application of these two methods to various available analytical and numerical results, which proves robustness of the procedure and also reveals the strong points and weaknesses. In chapter 4, conditions of molten ceramic droplets upon impact on the substrate, i.e. diameter, temperature, and speed, are calculated using a coupled Finite Volume-Discrete Phase simulation. A 3-dimensional Finite Volume simulation of a simplified plasma torch is used to track these particles during evaporation, melting, and breakup from injection port to the point of impact on the substrate. Various parameters that influence SPS process are studied utilizing the improved solution. At the injection port, parameters of interest are injection angle, velocity, and location. The effects of torch power and mass flow rate are also examined to ensure the quality of particles upon impact on the substrate are optimized. Moreover, special focus is dedicated to in-flight change of properties occurring for suspension droplets as they travel from injection site towards the substrate. Methods are used to update specific heat capacity, density, etc for
each YSZ-water droplet as evaporation makes the liquid content in each drop vanish. Also, different available models for prediction of suspension viscosities are reviewed and the suitable candidate for the ceramic suspension of interest here is chosen by examining them against available experimental data. Moreover, different parameters influencing final estimations of viscosity values such as maximum packing factors are studied. Chapter 5 presents the application of SPH model with phase change on study of impact and solidification of YSZ droplets generated during Suspension Plasma Spraying. Results are used for making predictions on the final coating of the substrate. And finally, chapter 6 concludes this thesis.
2.1 SPH Principles

Equations are discretized over a domain filled with SPH particles using integral interpolations. Consider the definition of the Dirac delta ($\delta$) function in the form of

$$\delta(x - x_i) = \begin{cases} \infty & x = x_i \\ 0 & x \neq x_i \end{cases}$$  \hspace{1cm} (2.1)

which has the identity property in the form of $\int_{-\infty}^{\infty} \delta(x - x_i) dx = 1$. For integral representation of a function like $f(x)$, it can easily be shown that

$$\int \phi f(x)\delta(x - x_i)dx = f(x_i)$$ \hspace{1cm} (2.2)

A more general case of this integral interpolation can be obtained by substituting delta function with an arbitrary smoothing kernel, $W(x - x_i, h)$, which has the following
two properties:
\[
\lim_{h \to 0} W(x - x_i) = \delta(x - x_i) \tag{2.3}
\]
\[
\int_{-\infty}^{\infty} W(x - x_i) dx = 1 \tag{2.4}
\]

\(h\), which is usually accompanied by an integer multiplicand, \(k\), determines the radius from \(x_i\) on which the smoothing function is non-zero. Therefore integral interpolation in general form is

\[
\int_{\phi} f(x) W(x - x_i) dx \cong f(x_i) \tag{2.5}
\]

which is only an approximation of \(f(x_i)\) unless \(W(x - x_i, h)\) is the Dirac delta function [44]. Using same methodology as of using Riemann sums, the integral above can be turned into a summation in the form of

\[
f(x_i) \cong -\sum_{j=1}^{N} \frac{m_j}{\rho_j} f(x_j) W(x_j - x_i, h) \tag{2.6}
\]

The gradient (or divergence) of \(f(x)\) can also be calculated using the same principles in the form of

\[
\nabla f(x_i) \cong \sum_{j=1}^{N} \frac{m_j}{\rho_j} f(x_j) \nabla_i W(x_j - x_i, h) \tag{2.7}
\]

It is however beneficial for numerical stability to use the following identities instead

\[
\nabla f(x) = \rho \left[ \nabla \left( \frac{f(x)}{\rho} \right) + \frac{f(x)}{\rho^2} \nabla \rho \right]. \tag{2.8}
\]
\[ \nabla f(x) = \frac{1}{\rho} [\nabla (\rho f(x)) - f(x) \nabla \rho] \]  

(2.9)

The SPH form of these identities after applying the same principles as mentioned before will be [44]

\[ \nabla f(x_i) = \rho_i \left[ \sum_j^N m_j \left( \frac{f(x_i)}{\rho_i^2} + \frac{f(x_j)}{\rho_j^2} \right) \nabla_i W_{ij} \right] \]  

(2.10)

and

\[ \nabla f(x_i) = \sum_j^N m_j \left[ \frac{f(x_j)}{\rho_j} - \frac{f(x_i)}{\rho_i} \right] \nabla_i W_{ij} \]  

(2.11)

### 2.2 Navier-Stokes equations

Navier-Stokes equations in a Lagrangian framework are

\[ \frac{D\rho}{Dt} = -\rho \nabla . v \]  

(2.12)

\[ \frac{Dv}{Dt} = \frac{1}{\rho} [-\nabla p + \mu \nabla^2 v + F^{st} + F^b] \]  

(2.13)

where \( F^b \) represents external body forces such as gravity and \( F^{st} \) is the surface tension force. These equations are closed by equation of state, which calculates pressure using density in the form of [45]

\[ P = P_0 (\frac{\rho}{\rho_0})^\gamma + b \]  

(2.14)

where \( \gamma = 7 \) and 1.4 for liquid and gas phases, respectively, \( b \) is a background pressure, and \( P_0 \) represents a reference pressure adjusted to keep maximum density deviations from \( \rho_0 \) in the order of \( O(1\%) \) [45].
Applying integral interpolation to continuity and Navier-Stokes equations

\[
\frac{D\rho_i}{Dt} \approx \sum_{j=1}^{N} m_j (\mathbf{v}_i - \mathbf{v}_j) \cdot \nabla_i W \tag{2.15}
\]

\[
\frac{D\mathbf{v}_i}{Dt} \approx -\sum_{j=1}^{N} m_j \left( \frac{p_i}{\rho_i^2} + \frac{p_j}{\rho_j^2} \right) \nabla_i W + \mathbf{F}^b + 4m_j \left( \frac{(\mu_i + \mu_j) \mathbf{x}_{ij} \cdot \nabla_i W}{(\rho_i + \rho_j)^2 |\mathbf{x}_{ij}|^2} \right) \mathbf{v}_i + \frac{\mathbf{F}_{ij}^{st}}{m_i} \tag{2.16}
\]

where \( m \) is the mass of each particle. To avoid possible singularities, it is beneficial to expand \( \frac{\mathbf{x}_{ij} \cdot \nabla_i W}{|\mathbf{x}_{ij}|^2} \) before calculations and remove a position vector that appears in both denominator and numerator, which can become close to or equal to zero.

Surface tension is calculated by applying interaction forces among particles in the form of [46]

\[
\mathbf{F}_{ij} = \begin{cases} 
S_{ij} \cos \left( \frac{15\pi}{3h} |\mathbf{x}_{ij}| \right) \frac{\mathbf{x}_{ij}}{|\mathbf{x}_{ij}|} & |\mathbf{x}_{ij}| \leq h \\
0 & |\mathbf{x}_{ij}| > h 
\end{cases} \tag{2.17}
\]

between each \( i \) and \( j \) particles. \( S_{ij} \) is a constant that controls the magnitude of surface tension force between each phase. Contact angles and interactions between fluids and solids can be obtained by choosing different values for \( S_{ij} \).

### 2.3 Particle Tracking

Tracking fluid surface is a challenging task in multiphase CFD studies. Knowledge of fluid interface helps in identifying where each phase is located inside the domain. Position and shape of interface can be used to calculate interfacial forces and phenomena such as surface tension, evaporation, chemical reactions, etc. Different CFD schemes have been used in tracking fluid interface. In Volume of Fluid Method (VOF), free surface is tracked by advection of a fraction function (\( C \)). This fraction function has
a value of 1 at one phase, 0 at the other, and a value between 0 and 1 at the interface. The value of this function is then advected through cells on domain based on fluid velocity vectors by

$$\frac{\partial C}{\partial t} + v \cdot \nabla C = 0$$

(2.18)

A similar methodology is also used in Level Set method. Here a function like $\phi$ is defined to divide the domain to three regions. $\phi$ gets the value of 0 on the fluid surface, has positive values on one side, and negative values on the other side of the interface. This curve is then moved using the same methodology as VOF by solving

$$\frac{\partial \phi}{\partial t} = v |\nabla \phi|$$

(2.19)

One of disadvantages of level set is the fact that it cannot conserve mass [47]. Both level set and VOF also need to have the equations mentioned before solved on the fluid domain which can be computationally expensive. SPH, being a Lagrangian method can help in removing this problem. Position of each SPH particle is updated at each iteration using

$$\frac{Dx}{Dt} = v$$

(2.20)

This makes sure that all particles, including those which form the interface, are moving with the fluid velocity. This procedure not only conserves mass exactly, but also rules out the need for solving any additional advection equation. In this method, particles of each phase are only needed to be marked once and the exact position of each phase will be available at any time due to the movement of the particles.
2.4 Boundaries

For particles that come in contact with a wall, the no-slip boundary condition needs to be applied. A proper implementation of this condition is of more importance in the scope of current work, as in most cases studied later, the fluid comes in contact with wall at comparably large velocities. Morris et al. [4] suggested giving the stationary wall particles an artificial velocity in the opposite direction of fluid flow. In this method, the distance of each particle from the wall needs to be calculated. Assuming the distance of a fluid particle outside the wall and the distance of a solid particle inside the wall are given as $d_{\text{fluid}}$ and $d_{\text{solid}}$, the artificial velocity vector for solid particles can be calculated from

$$V_{\text{solid}} = -\frac{d_{\text{solid}}}{d_{\text{fluid}}} V_{\text{fluid}}$$  \hspace{1cm} (2.21)

To avoid large velocities when particles are too close to the walls, it is customary to limit the value of $\frac{d_{\text{solid}}}{d_{\text{fluid}}}$ by 0.5. The main disadvantage of this formulation is that the exact location of the wall needs to be known prior to calculation. For instance, for cases with curved or vertical walls, or with moving boundaries such as a growing solidification front, the implementation of this algorithm is inefficient. To overcome this, Holmes et al. [48] have proposed an alternative method. For each particle $i$, a state-specific particle density is defined as

$$\bar{n}_i = \sum_j \delta W(x_{ij}, h)$$  \hspace{1cm} (2.22)

with $\delta$ being 1 when particles $i$ and $j$ are of the same state (both fluid or both solid), and 0 otherwise. In addition, a second particle density is defined which now contains all solid and fluid particles in the neighborhood of particle $i$.
\[ n_i = \sum W(x_{ij}, h) \]  \hspace{1cm} (2.23)

It is evident that the ratio of \( \bar{n}_i \) and \( n_i \), \( \chi = \frac{\bar{n}_i}{n_i} \), is inversely proportional to the number of particles from the other phase existing in the neighborhood of particle \( i \).

For particles in the bulk of fluid (or solid), \( \chi_i = 1 \), for particles located exactly on the wall, \( \chi_i = 0.5 \), and for the rest \( \chi_i \) is between these two limits. Using this ratio, Holmes et al. [48] have shown that the artificial velocity of solid particles in the wall can be calculated from

\[ V_{\text{solid}} = -\frac{\zeta_{\text{solid}}}{\max \left( \zeta_{\text{fluid}}, \frac{\sqrt{3}}{4} h \right)} V_{\text{fluid}} \]  \hspace{1cm} (2.24)

with \( \zeta = \frac{kh}{0.5} (\chi - 0.5) \). This formulation can be used to satisfy the no-slip boundary condition on both flat and curved boundaries and hence has been used here.

Aside from the no-slip boundary condition, particles at high velocities may also penetrate the walls. To prevent this, a short-range repulsive force is placed on wall particles. This force is only active when the fluid particle is too close to a wall particle, say their distance is below initial particle spacing. This force is typically chosen in the form of Lennard-Jones potential as

\[ f(x) = \begin{cases} D \left[ \left( \frac{x_0}{|x^*|} \right)^{12} - \left( \frac{x_0}{|x^*|} \right)^{6} \right] \frac{x^*}{x_0^{12}} & |x^*| < x_0 \\ 0 & |x^*| \geq x_0 \end{cases} \]  \hspace{1cm} (2.25)

where \( D \) is a constant used for calibration, \( x^* \) is the position vector pointing from the boundary particle towards the target fluid particle, and \( x_0 \) is the cutoff distance, taken equal to the initial particle spacing here.
2.5 Tensile Instability

It is a widely known concept that SPH particles subjected to negative pressures show tendency of forming small clusters. While simulation results are typically not significantly affected in the larger scale from this arrangement, it becomes important for the test cases of interest here. Having large impact velocities leads to large displacements in particle positions. For a single droplet impact, as particles in the drop are typically arranged nicely before the impact, this is not problematic. However, when multiple particles are impacting one after another, the particles from previous impacts, that now are in near stationary condition might have formed clusters, that would cause instabilities after secondary impacts. Moreover, as the solidification front moves across the particles, solidification of a portion of particles in a cluster will make the solution unstable, as particles that prior to solidification were closer to one another and in equilibrium, will now be unbalanced and too close to solid particles to properly recover their positions. To overcome this, a proper model needs to be used to limit particle clusterings. Tensile instabilities have been studied in detail by studies such as Melen et al. [49] and Gray et al. [50]. Treatments offered by these studies, while being effective, are computationally costly. It was found here that more simple solutions, such as the model of Monaghan [51], also worked well in the current framework. In this algorithm, a correction term is added to the momentum equation which adjusts particles when negative pressures are present in the form of

\[ \Pi = R^* f_{ij}^n \]  \hspace{1cm} (2.26)

Here, \( f_{ij} = \frac{W(x_{ij}, h)}{W(x_0, h)} \). \( x_0 \) is the initial spacing between particles, and \( n \) has the value of 4. The factor, \( R^* \), is written as
\[ R^* = R^*_i + R^*_j \]  

\[ R_i = \begin{cases} \epsilon \frac{|p_i|}{\rho_i^2} & \text{if } p_i < 0 \\ 0 & \text{otherwise} \end{cases} \]  

where the value for \( R_i \) and \( R_j \) is calculated from

2.6 Time Marching

In the current scope, particle positions and the rest of variables are updated at each time step using the conventional SPH formulation in the form of

\[ \frac{Dx_i}{Dt} = V_i \]  

Algorithms such as velocity-verlet, Leapfrog, predictor-corrector, or Runge-Kutta can be used for performing numerical integration. The Leapfrog method has been chosen here. As explicit time marching schemes are stable under Courant-Friedrichs-Lewy (CFL) condition, time steps are limited here using

\[ \Delta t \leq 0.25 \frac{h}{c} \]  

where \( c \) is the speed of sound. Also based on viscous diffusion, another constraint can be imposed in the form of
\[ \Delta t \leq 0.125 \frac{h^2}{\nu_{max}} \]  
\hspace{1cm} (2.32)

with \( \nu \) being the maximum kinematic viscosity in the domain. For when acceleration forces are large, it is also beneficial to write a constraint based on the magnitude of forces exerted on each particle (\( f \))

\[ \Delta t \leq 0.125 \left( \frac{h}{f} \right)^{0.5} \]  
\hspace{1cm} (2.33)

The value of time steps are also limited by the choice of neighbour search algorithm. Depending on how particle neighbours are updated, time steps should be limited to prevent a particle from escaping or entering a neighbourhood without getting noticed.

### 2.7 Latent heat evaluation

#### 2.7.1 Enthalpy Formulation with phase-change

**Energy equation**

The energy equation here is solved using methodology proposed by Cao et al. [52]. The three dimensional energy equation for laminar flow with no viscous dissipation is

\[ \frac{\partial}{\partial t} (\rho H) + \frac{\partial}{\partial x} (\rho u H) + \frac{\partial}{\partial y} (\rho v H) + \frac{\partial}{\partial z} (\rho w H) = \frac{\partial}{\partial x} \left( k \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( k \frac{\partial T}{\partial y} \right) + \frac{\partial}{\partial z} \left( k \frac{\partial T}{\partial z} \right) \]  
\hspace{1cm} (2.34)

For an incompressible fluid,

\[ \frac{dH}{dT} = C(T) \]  
\hspace{1cm} (2.35)
Enthalpy is calculated with respect to a reference point. In the current context, the reference point is chosen to be the melting point of the solid material. At this temperature, enthalpy is assumed to be zero (see figure 2.1).

\begin{equation}
T = \begin{cases}
T_m + H/C_s & H \leq 0 \\
T_m & 0 < H < L \\
T_m + (H - L)/C_l & H \geq L
\end{cases}
\end{equation}

and for materials like alloys where phase change occurs over a temperature range,
\[ T - T_1 = \begin{cases} 
\frac{H}{C_s} & H \leq 0 \quad \text{solid} \\
\frac{\Delta T H}{L + C_m \Delta T} & 0 < H < L + C_m \Delta T \quad \text{mushy} \\
\frac{H}{C_l} - \frac{[L + (C_m - C_l) \Delta T]}{C_l} & H \geq L + C_m \Delta T \quad \text{liquid}
\end{cases} \quad (2.37) \]

As shown in figure 2.1, for materials where phase change occurs at a single temperature, enthalpy experiences a step jump at the melting temperature \( T_m \). For materials such as alloys with a phase change temperature interval (PCTI), phase change occurs over a temperature range \( T_1 \) to \( T_2 \). When the temperature is within the PCTI, the material is mushy. In the current study and equation 2.37, enthalpy is assumed to vary linearly over the PCTI.

**Kirchhoff temperature**

In order to eliminate the constant values in equations 2.36 and 2.37, the Kirchhoff temperature is defined as

\[ T^* = \int_{T_{ref}}^{T} k(\zeta) d\zeta \]

Applying this definition to equations 2.36 and 2.37 gives

\[ T^* = \begin{cases} 
k_s(T - T_m) & T < T_m \\
0 & T = T_m \\
k_l(T - T_m) & T > T_m
\end{cases} \quad (2.38) \]

for a material with a single phase change temperature and

\[ T^* = \begin{cases} 
k_s(T - T_1) & T \leq T_1 \\
k_m(T - T_1) & T_1 < T < T_2 \\
k_l(T - T_1) & T \geq T_2
\end{cases} \quad (2.39) \]

for a material that changes phase over a temperature interval.
The constant values $T_m$ and $T_1$ can now be omitted by substituting $T$ from equations 2.36 and 2.37 into equations 2.38 and 2.39, respectively. This gives the Kirchhoff temperature as a function of enthalpy in the form of

$$ T^* = \begin{cases} 
  k_s H / C_s & H \leq 0 \\
  0 & 0 < H < L \\
  k_l (H - L) / C_l & H \geq L 
\end{cases} \quad (2.40) $$

for a single phase change temperature and

$$ T^* = \begin{cases} 
  k_s H / C_s & H \leq 0 \\
  k_m [\Delta TH / (L + C_m \Delta T)] & 0 < H < L + C_m \Delta T \\
  k_l [H / C_l - (L + (C_m - C_l) \Delta T) / C_l] & H \geq L + C_m \Delta T 
\end{cases} \quad (2.41) $$

for phase change over a temperature range. The linearity of equations 2.40 and 2.41 allows the calculation of $H$ or $T^*$ when only one is known.

**Source terms**

Expressing $T^* = \Gamma(H) H + S(H)$, $\Gamma$ and $S$ can be derived from equations 2.40 and 2.41 as

$$ \Gamma(H) = \begin{cases} 
  k_s / C_s & H \leq 0 \\
  0 & 0 < H < L \\
  k_l / C_l & H \geq L 
\end{cases} \quad (2.42) $$

$$ S(H) = \begin{cases} 
  0 & H \leq 0 \\
  0 & 0 < H < L \\
  -Lk_l / C_l & H \geq L 
\end{cases} $$

for a single temperature phase change and
\( \Gamma(H) = \begin{cases} 
\frac{k_s}{C_s} & H \leq 0 \\
\frac{k_m \Delta T}{(L + C_m \Delta T)} & 0 < H < L + C_m \Delta T \\
\frac{k_l}{C_l} & H \geq L + C_m \Delta T 
\end{cases} \) 

\( S(H) = \begin{cases} 
0 & H \leq 0 \\
0 & 0 < H < L + C_m \Delta T \\
k_l \frac{[L + (C_m - C_l) \Delta T]}{C_l} & H \geq L + C_m \Delta T 
\end{cases} \) 

(2.43)

for phase change over a temperature range.

**Reconstruction of Lagrangian energy equation**

With the definition of \( T^* \), equation 2.34 can be rewritten as

\[
\frac{\partial}{\partial t} (\rho H) + \frac{\partial}{\partial x} (\rho u H) + \frac{\partial}{\partial y} (\rho v H) + \frac{\partial}{\partial z} (\rho w H) = \frac{\partial^2 T^*}{\partial x^2} + \frac{\partial^2 T^*}{\partial y^2} + \frac{\partial^2 T^*}{\partial z^2} 
\]

(2.44)

and further expansion of \( T^* \) using equations 2.42 or 2.43 transforms equation 2.44 into

\[
\frac{\partial}{\partial t} (\rho H) + \frac{\partial}{\partial x} (\rho u H) + \frac{\partial}{\partial y} (\rho v H) + \frac{\partial}{\partial z} (\rho w H) = \frac{\partial^2 (\Gamma H)}{\partial x^2} + \frac{\partial^2 (\Gamma H)}{\partial y^2} + \frac{\partial^2 (\Gamma H)}{\partial z^2} + \frac{\partial^2 S}{\partial x^2} + \frac{\partial^2 S}{\partial y^2} + \frac{\partial^2 S}{\partial z^2} 
\]

(2.45)

For simplicity of notations, equation 2.44 can be rewritten as

\[
\frac{\partial}{\partial t} (\rho H) + \nabla \cdot (\rho H \mathbf{V}) = \nabla^2 T^* 
\]

(2.46)

By expanding \( \nabla \cdot (\rho H \mathbf{V}) \) into
\[
\frac{\partial}{\partial t}(\rho H) + \mathbf{V} \cdot \nabla (\rho H) + (\rho H) \nabla \cdot \mathbf{V} = \nabla^2 T^* \tag{2.47}
\]

the first two terms on the left hand side can be combined to form a total derivative,

\[
\frac{D(\rho H)}{Dt} = -(\rho H) \nabla \cdot \mathbf{V} + \nabla^2 T^* \tag{2.48}
\]

Equation 2.48 for the total derivative is the Lagrangian form of the transformed enthalpy equation. This equation gives the variation of \( \rho H \) with time for each SPH particle. Using the Lagrangian continuity equation \( \frac{D\rho}{Dt} + \rho \nabla \cdot \mathbf{V} = 0 \), the term \( -\rho \nabla \cdot \mathbf{V} \) in equation 2.48 can be replaced by the total derivative of density resulting in

\[
\frac{D(\rho H)}{Dt} = H \frac{D\rho}{Dt} + \nabla^2 T^* \tag{2.49}
\]

**SPH discretization**

The enthalpy equation, in the form of equation 2.49, is discretized using an SPH formulation here. This equation resolves enthalpy while applying the effects of phase change latent heat. Here, \( \rho H \) is taken as a single variable. At each iteration, the variation of \( \rho H \) with respect to time is calculated and is used to obtain a new value for \( \rho H \) for each particle in an explicit manner. Wherever a value of \( H \) is needed, \( \rho H \) for that particle is divided by its density. In this case, equation 2.49 for particle \( i \) is written as

\[
\frac{D(\rho H)_i}{Dt} = H_i \frac{D\rho_i}{Dt} + (\nabla^2 T^*)_i = H_i \frac{D\rho_i}{Dt} + (\nabla^2 \Gamma H)_i + (\nabla^2 S)_i \tag{2.50}
\]

In equation 2.50, the values for \( H_i \) and \( D\rho_i/Dt \) are known for each particle. The Laplacians for \( \Gamma H \) and \( S \) must be discretized and calculated in SPH form. To ensure
numerical stability, these Laplacians are broken into two steps in the form of

\[(\nabla^2 \Gamma H)_i + (\nabla^2 S)_i = \nabla_i \cdot (\nabla_i (\Gamma H) + \nabla_i S)\]  \hspace{1cm} (2.51)

The inner gradient is calculated here using equation 2.52 while the outer divergence is calculated using equation 2.53

\[
\nabla f(x_i) = \sum_j^N \frac{m_j}{\rho_i} [f(x_j) - f(x_i)] \cdot \nabla_i W_{ij} \hspace{1cm} (2.52)
\]

\[
\nabla \cdot \Phi(x_i) = \rho_i \left[ \sum_j^N m_j \left( \frac{\Phi(x_i)}{\rho_i^2} + \frac{\Phi(x_j)}{\rho_j^2} \right) \cdot \nabla_i W_{ij} \right] \hspace{1cm} (2.53)
\]

In this manner, the inner gradient in equation 2.50 becomes

\[
G_i = \nabla_i (\Gamma H) + \nabla_i S = \sum_j^N \frac{m_j}{\rho_i} \{[\Gamma(H_j)H_j + S(H_j)] - [\Gamma(H_i)H_i + S(H_i)]\} \nabla_i W_{ij} \hspace{1cm} (2.54)
\]

Here, the values for \(\Gamma\) and \(S\) are calculated using equations 2.42 and 2.43 using the values of \(H\) at the particles \(i\) and \(j\). Then the outer divergence in equation 2.51 becomes

\[
(\nabla \cdot G)_i = \rho_i \left[ \sum_j^N m_j \left( \frac{G_i}{\rho_i^2} + \frac{G_j}{\rho_j^2} \right) \cdot \nabla_i W_{ij} \right] \hspace{1cm} (2.55)
\]

These discretizations, however, can become unstable in some cases, especially when there is a discontinuity in the physical properties in the domain [53], for example, heat transfer between two materials with different thermal conductivities. To overcome this issue, a remedy suggested by Cleary et al. [53] has been used here. With this new formulation, the Laplacians in equation 2.50 are directly calculated using
\[ \nabla^2(\Gamma H + S)_i = \sum_{j}^{N} 2 \frac{m_j}{\rho_j} (f_i - f_j) \frac{\nabla_i W_{ij}}{r_{ij}} \]  

(2.56)

where the variable \( f \) is defined as

\[ f_i = \Gamma_{eff} H_i + S_i \]
\[ f_j = \Gamma_{eff} H_j + S_j \]  

(2.57)

and \( \Gamma_{eff} \) is defined as

\[ \Gamma_{eff} = \begin{cases} 
\frac{2\Gamma_i\Gamma_j}{\Gamma_i + \Gamma_j} & \Gamma \neq 0 \\
0 & \text{otherwise} 
\end{cases} \]  

(2.58)

Substituting back into equation 2.50, the variations of \( \rho H \) for particle \( i \) is calculated from

\[ \frac{D(\rho H)_i}{Dt} = H_i \left( \frac{D\rho}{Dt} \right)_i + \nabla^2(\Gamma H + S)_i \]  

(2.59)

### 2.7.2 Effective heat capacity

Transient heat conduction is mathematically represented as

\[ \frac{\partial H}{\partial t} = \nabla \cdot (k\nabla T) \]  

(2.60)

By expressing \( H \) as a function of \( T \), equation 2.60 can be written as

\[ \frac{dH}{dT} \frac{\partial T}{\partial t} = \nabla \cdot (k\nabla T) \]  

(2.61)

In this form, equation 2.61 does not account for the release (absorption) of latent heat during solidification (melting). To do that, the parameter
$C = \frac{dH}{dT}$ (2.62)

known as the effective heat capacity, can be modified to include the effect of latent heat, as follows [54,55]

$$C = \begin{cases} 
  C_s & T < T_m \\
  C_m + L\delta(T - T_m) & T = T_m \\
  C_l & T > T_m 
\end{cases}$$ (2.63)

where $\delta(T - T_m)$ is the Dirac delta function. It can be easily confirmed that using this form of $C$ yields a total heat release of $\int_{-\infty}^{\infty} L\delta(T - T_m)dT = L$.

First, SPH discretizations of two finite element methods for solving equation 2.60 are presented. Then, by starting with a simple approximation, new SPH interpolations for the inclusion of latent heat via equation 2.63 are derived. This leads to five different methods, that are presented in this paper as equations 2.65, 2.66, 2.72, 2.73, and 2.76.

**Variational Methods**

A first approach is to use the variation of enthalpy as a function of temperature to calculate the value of $C$ using equation 2.62. The first equation presented here was developed by Del Diudice et al. [56] by transforming equation 2.62 using the direction of the temperature gradient, $s$, as [23]

$$C = \frac{\partial H/\partial s}{\partial T/\partial s} = \frac{(\partial H/\partial x)l_{sx} + (\partial H/\partial y)l_{sy} + (\partial H/\partial z)l_{sz}}{\partial T/\partial s}$$ (2.64)

with direction cosines defined as $l_{s\alpha} = \frac{\partial T}{\partial \alpha}/\partial \alpha$ and $\frac{\partial T}{\partial s} = \sqrt{(\frac{\partial T}{\partial x})^2 + (\frac{\partial T}{\partial y})^2 + (\frac{\partial T}{\partial z})^2}$.

This gives an effective heat capacity
Chapter 2. Mathematical Formulation and Numerical Model

\[ C = \frac{(\partial H/\partial x)(\partial T/\partial x) + (\partial H/\partial y)(\partial T/\partial y) + (\partial H/\partial z)(\partial T/\partial z)}{(\partial T/\partial x)^2 + (\partial T/\partial y)^2 + (\partial T/\partial z)^2} \]  

(2.65)

Another approach is similar to that of Lemmon [57], where \( s \) is taken as the normal direction of the interface, and yields

\[ C = \left[ \frac{(\partial H/\partial x)^2 + (\partial H/\partial y)^2 + (\partial H/\partial z)^2}{(\partial T/\partial x)^2 + (\partial T/\partial y)^2 + (\partial T/\partial z)^2} \right]^{1/2} \]  

(2.66)

These two equations were implemented in a finite element context by Thomas et al. [21] and Dalhuijsen et al. [23]. To discretize equations 2.65 and 2.66 for SPH, an approach inspired by Del Giudice et al. [56] is used here. In their finite element method, the enthalpy is smoothed by a shape function before using the above equations. Here, the value of enthalpy for each SPH particle is calculated from its temperature using [23]

\[
C = \begin{cases} 
\int_{T_{ref}}^T \rho C_s(T)dT & T < T_1 \\
\int_{T_{ref}}^{T_1} \rho C_s(T)dT + \int_{T_1}^T \rho (dL/dT + C_m(T))dT & T_1 \leq T \leq T_2 \\
\int_{T_{ref}}^{T_1} \rho C_s(T)dT + \rho L + \int_{T_1}^{T_2} \rho C_m(T)dT + \int_{T_2}^T \rho C_i(T)dT & T > T_2 
\end{cases}
\]  

(2.67)

To increase accuracy and stability, we propose that the value of enthalpy at each particle is smoothed using

\[ H_i^s = \sum_j \frac{m_j}{\rho_j} H_j W_P(x_j - x_i, h) \]  

(2.68)

With the value of enthalpy known, the gradients of enthalpy that appear in equations 2.65 and 2.66 can be calculated using
(\nabla H)_i = \sum_j m_j \frac{m_j}{\rho_j} \left[H^*_j - H^*_i\right] \nabla W^P(x_j - x_i, h) \quad (2.69)

A similar formulation is used to calculate the first gradient of temperature. The second gradient of temperature that appears in these equations is calculated by taking the gradient of the first gradient:

\begin{align*}
(\nabla^2 T)_i &= \rho_i \sum_j m_j \left[ \frac{\nabla T_j}{\rho_j^2} + \frac{\nabla T_i}{\rho_i^2} \right] \cdot \nabla W^P(x_j - x_i, h) \quad (2.70)
\end{align*}

**Step Release**

Calculation of the partial derivatives appearing in equations 2.65 and 2.66 demands many computational steps, and so motivates the search for alternative methods. Returning to equation 2.63, it is convenient to replace $\delta$, that releases latent heat at a single temperature $T_m$, with a function that releases the latent heat over a finite temperature interval around $T_m$. Hsiao [22] shows that a very simple substitute for $\delta$ is of the form

\begin{align*}
C &= \begin{cases} 
C_s & T < T_m - \Delta T \\
C_m + \frac{L}{2\Delta T} & T_m - \Delta T \leq T \leq T_m + \Delta T \\
C_l & T > T_m + \Delta T \end{cases} \quad (2.71)
\end{align*}

Here the total latent heat released over the interval of $2\Delta T$ is still $L$. Figure 2.2 shows this step jump in the apparent heat capacity.

The main drawback of this approach is the need for a phase change temperature interval. For some particles during a simulation, the value of temperature may jump from a value above this interval to a value below it in one time step [22]. This means that the effect of latent heat might be bypassed if the temperature variation in a time
step is large compared to $\Delta T$, and imposes a limitation on the solution time step [23]. Additionally, for the case of pure materials where solidification/melting occurs at a single temperature, the artificial phase change interval [21] has no physical meaning. A poor choice of this interval may cause solution inaccuracy.

For the phase change of alloys with a mushy zone, where solidification/melting occurs over a temperature range, equation 2.71 becomes

$$C = \begin{cases} 
  C_s & T \leq T_1 \\
  C_m + \frac{L}{T_2-T_1} (T_1 < T < T_2) \\
  C_l & T \geq T_2 
\end{cases} \quad (2.72)$$

![Figure 2.2: Apparent heat capacity with a step jump for the latent heat.](image)

**Gradual Release**

A second approach is to replace the Dirac delta function $\delta$ by an even smoothing function $W$ that has the identity property of $\delta$, $\int_{-\infty}^{\infty} W(T - T_m, h^*)dT = 1$, and is defined over a temperature range, shown here as $\Delta T = k^* \times h^*$. The integer $k^*$ and the float $h^*$ are similar in definition to the usual SPH notations of $k$ and $h$, the
only difference being that $k$ and $h$ are defined in the space domain and are based on particle positioning, while $k^*$ and $h^*$ are defined over the temperature domain of particles. In other words, where the value of $k \times h$ determines a radius around each particle within which its neighbours contribute to a heat transfer calculation, similarly, $k^* \times h^*$ defines a radius in the one dimensional domain of temperature around the melting temperature. This new radius acts as a second filter in addition to $k \times h$. In formulations where both filters are present, the only particles that contribute to heat transfer are those close to a certain particle in both the space and temperature domains.

Replacing $\delta$ with the smoothing function $W$, depending on the choice of $W$, has been shown to have a second or higher order of accuracy [44]. Figure 2.3 demonstrates the smoothing function. More detailed discussions on the derivation of a SPH formulation, and more general integral interpolations, can be found in [58,59]. Using this definition, equation 2.63 can be rewritten as

$$C = \begin{cases} 
C_s & T < T_m - \Delta T \\
C_m + LW^T(T - T_m, h^*) & T_m - \Delta T \leq T \leq T_m + \Delta T \\
C_i & T > T_m + \Delta T 
\end{cases} \quad (2.73)$$

where $W^T$ is a 1D kernel defined in the temperature domain.

**Smoothed Gradual Release**

The accuracy and efficiency of equation 2.73 can be further improved by smoothing the term $LW^T(T - T_m, h^*)$ with a higher order kernel [60,61]. This is achieved using the standard SPH interpolation [13]
Chapter 2. Mathematical Formulation and Numerical Model

\[ f(x_0) \approx -\sum_{j=1}^{N} \frac{m_j}{\rho_j} f(x_j) W(x_i - x_0, h) \]  
\hspace{1cm} (2.74)

Figure 2.3: The smoothing function \( W \).

Summation here is performed over all \( j \) particles located in the neighbourhood radius of particle \( i \). With this definition, the latent heat in equation 2.73 at the position of an arbitrary particle \( i \) is smoothed using

\[ L_i^s = \sum_{j=1}^{N} \frac{m_j}{\rho_j} [L W^T(T_j - T_m, h^*)] W^P(x_j - x_i, h) \]  
\hspace{1cm} (2.75)

and hence equation 2.63 can be rewritten as

\[ C = \begin{cases} 
C_s & T < T_m - \Delta T \\
C_m + L^s & T_m - \Delta T \leq T \leq T_m + \Delta T \\
C_l & T > T_m + \Delta T 
\end{cases} \]  
\hspace{1cm} (2.76)

\( \Delta T \) is identical to the radius of influence of the smoothing function, \( W^T(T - T_m, h^*) \).

In this formulation, the latent heat is distributed to particles based on the difference between the particle temperature and the melting temperature, and also based on how
far other particles in its neighbourhood are from the reference melting temperature. In other words, this equation accounts for two variations: 1) changes in temperature of each particle with respect to the melting point, and 2) the variation of temperature in the domain with respect to the position vector $\mathbf{x}$. The term $L W^T(T_j - T_m, h^*)$ is a discretization of $L\delta$ and a function of temperature only. Furthermore, temperature itself is a function of particle position in the domain $T = T(x)$. The second term, $W^P(x_j - x_i, h)$, accounts for the change of temperature with respect to position. The smoothing behaviour of equation 2.75 assures there is no sudden jump in enthalpy within the domain with respect to changes in the position vector $\mathbf{x}$.

### 2.7.3 Mushy zone handling

As discussed before for the case of alloys, phase change goes through a mushy zone during melting and solidification. The existence of this region is due to the difference in phase change temperatures of elements forming the alloy. One element might start the solidification process sooner and along the way, the solid crystals that will nucleate and grow will lead to a slurry of solid and molten materials. This phenomena might happen in various ways. In one, the solidified phases might be growing at discrete sites. In this manner, the physical properties of the mixture will be closer to those of a suspension of solid phases in a liquid phase. On the other hand, the solidification might be happening as a solid front growing from a wall. This solid front will have smaller scale porous branches that spread into the heart of the still molten phase. In this manner, the liquid phase will be flowing through the porous solid phase that is growing in size. The flow in this case will be close to flow of liquid phase through porous material. Detailed explanations of these conditions can be found in the work of Voller et al. [62].
The effect of the mushy region is typically included in one of two ways. When the flow pattern is similar to flow through porous media, a Darcy source term can be added to the momentum equation, to account for the pressure drop for the fluid moving through pores. This flow is usually found in casting problems [62]. When the flow is more homogeneous, the effect of having a slurry-like mixture is accounted for by modifying the viscosity. In the current study, this approach has been chosen as it avoids variations in pressure forces that can lead to instabilities in SPH. Depending on the temperature or enthalpy values, a liquid fraction $\alpha$ and a solid fraction $\beta = 1 - \alpha$ can be defined for each particle at each iteration. Based on these values, the viscosity of each particle is updated using [62]

$$\mu = \alpha \mu_{\text{liquid}} + \beta \mu_{\text{solid}}$$ (2.77)

For particles that have a liquid fraction of unity, a new viscosity will be equal to the viscosity of the molten material. For liquid fractions of zero, the viscosity of each particle is set to very large to mimic solid matter. In between these two limits, viscosity is varied linearly. Some researchers have substituted this linear variation with [63]

$$\mu = \mu_{\text{liquid}} \left( 1 + C \frac{\beta^3}{(1 - \beta)^3 + \epsilon} \right)$$ (2.78)

where $C$ is a problem dependent constant and $\epsilon$ is introduced to avoid division by zero. The values for viscosity need to be calibrated for different materials and conditions. As will be shown later, equation 2.77 provides more stable solutions in the current framework and hence has been utilized in this study.
Chapter 3

Validation

Before utilizing the SPH methods discussed so far to study the impact and solidification of molten ceramic droplets generated by SPS, it is appropriate to have them validated. In the following chapter, the SPH methods discussed before have been validated. Initially the surface tension method is tested for its ability in formation of droplets and capturing pressure jump across the surface of these droplets. It has to be noted at this point that though the surface tension module is capable of reconstructing capillary effects, as it will be shown later, these effects are minimal in most stages of the problems of interest here. Secondly, the phase change methods have been tested against multiple analytical and numerical data. Finally, based on the results, the method more suited for the problems of interest here has been chosen for studies that follow in the rest of the chapters.

3.1 Surface Tension

Surface tension in the form discussed before is capable of capturing accurate pressure jump across the droplet’s surface. Moreover, small number of total particles con-
tribute to the surface tension calculations, which makes the solution more efficient. Figure 3.1 shows how surface tension generates a spherical drop from an initial cubic positioning. The radius of the drop is 300µm and the fluid under study is very similar to water with a density of 1000kg/m³, viscosity of 1 × 10⁻³kg/ms, and a surface tension coefficient of 90mN/m. Evolution of shape of the drop during this formation has been demonstrated in this figure.

During calculations, local pressure at each particle is determined using equation of state in the form of \( P = P_0 \frac{\rho}{\rho_0} \gamma + b \). For pressure jump across the drop’s interface however, this equation is not reliable for small size drops [46]. Since surface tension is imposed using particle-particle interactions and droplet sizes are small, methods available in Molecular Dynamics can be used in estimation of the pressure inside the droplet. For a drop at equilibrium, pressure can be estimated using

\[
P = S^{-1} \sum_k f_k
\]

(3.1)

where \( S \) is the area of spherical surface \( (4\pi r^2) \) with a radius of \( r \), known as the viral radius [64]. \( f_k \) is the normal components of pair forces acting on the surface. Figure 3.2 shows the pressure calculated inside a drop with a radius of \( 2.9 \times 10^{-4}m \) filled with 15625 SPH particles with a density of 1000kg/m³ and viscosity of \( 1 \times 10^{-3}Pa\cdot s \).

The pressure inside the drop, as reported by Tartakovsky [46], can be estimated by averaging over the middle area of this chart. The initial peak in this chart can be explained by noting that these points are located in a radius which is in the order of mesh resolution \( (2 \times 10^{-5} \text{ here}) \). This means that not enough particles will be available in this search area to have surface tension and pressure forces balanced out.

With the pressure estimated, the surface tension coefficient can now be determined using the Young-Laplace equation. Figure 3.3 shows the surface tension coefficient
(\sigma) that has been calculated inside drops with different diameters (mesh resolution is the same for all these cases).

Figure 3.4 shows pressure variations inside a drop with a constant radius of $2.8 \times 10^{-4} m$. Mesh resolution is the parameter changing here going from $30 \times 30 \times 30$ particles inside the drop to $8 \times 8 \times 8$ particles. This figure suggests that pressure variations inside drop converge to uniform values as finer mesh resolutions are used. Figure 3.5 shows pressure jump at equilibrium for when particles are initially placed in the form of a droplet with a lower surface tension magnitude. $PPD$ here represents the number of particles placed on the droplet’s diameter and has the values of 10, 20, 40, and 80. Using this surface tension model, capillary effects can also be captured. These effects are not of particular interest in the studies that follow here and have been included for demonstration purposes only. As an example, the contact angle that a drop makes on a solid surface can also be captured by assigning different interaction forces between solid and fluid. Two test cases have been demonstrated in figure 3.6 showing water drops spreading on a surface under gravitational force of $9.81 m/s^2$. Drops are initially left on the surface with no velocity and in the form of a cube. Contact angles here are measured at $80^\circ$ and $160^\circ$. 
Figure 3.1: Formation of a 3D drop under surface tension from initial cubic shape
Figure 3.2: Calculated pressure jump of the drop at equilibrium

Figure 3.3: Surface tension coefficient ($\sigma$) for drops with different diameters.
Figure 3.4: Calculated pressure jump of drops at equilibrium for different mesh resolutions.

Figure 3.5: Calculated pressure jump of the drop at equilibrium. Particles are initially arranged in the form of a droplet. $PPD$ represents particles per diameter.
Figure 3.6: Water drops spreading on a surface with a constant contact angle. Contact angles are 80° and 160°.
3.2 Phase-Change: Enthalpy Formulation

3.2.1 Conduction Problem

Comparison to Analytical Solution

Similar to Cleary et al. [53], a two-dimensional infinite slab is assumed in $x$–$y$ plane with a temperature discontinuity in its middle at $x = 0$. The material properties are $\rho_1, k_1, C_1$ when $x < 0$ and $\rho_2, k_2, C_2$ when $x > 0$. The transient analytical solution to this problem with an initial temperature jump of $T_0$ across the discontinuity is in the form of [65]

$$T - T_{01} = \begin{cases} \frac{k_1}{k_1^{1/2} + k_2^{1/2} \alpha} \left[ 1 + \frac{k_2}{k_1} \alpha^{1/2} \text{erf} \left( \frac{x}{2\sqrt{\alpha t}} \right) \right] & x > 0 \\ \frac{k_1}{k_1^{1/2} + k_2^{1/2} \alpha} \left[ \text{erfc} \left( \frac{|x|}{2\sqrt{\alpha t}} \right) \right] & x < 0 \end{cases}$$

(3.2)

Here, $\alpha = k/\rho C$ is the thermal diffusivity. To avoid the complications of modeling an infinite slab, the problem is modeled here with slight modifications [53]. To make the heat transfer one dimensional, a periodic boundary condition is applied perpendicular to heat flow direction in $y$ direction. The $x$ axis is also limited to $-1 < x < 1$ and a no-heat flux boundary condition is applied to it ends. It is evident the temperature profile will be slightly different than the analytical solution near the $x$ limits, especially as time goes to infinity. The value of the temperature across all points should converge to $T_0/2$ by the end. These test cases are conducted with 80 particles in $x$ direction. For the first case, the temperature jump and all thermal properties are taken to be unity. Figure 3.7 shows the temperature profile for this case for 0, 1, 3, 8, 10, and 20$\mu$s. The solid lines in this figure are the analytical solutions obtained from equation 3.2. As expected, temperature values near the $x$ limits of $-1$ and $1$ start deviating from the analytical solution as time becomes larger. The values of temperature in
the middle of the slab however are a good match to the analytical solution.

![Temperature profile across the slab with an initial unit temperature jump at $x = 0$.](image)

Figure 3.7: Temperature profile across the slab with an initial unit temperature jump at $x = 0$.

The second case shows performance under discontinuity in thermal properties. The temperature jump is taken to be 10 and thermal properties of materials are same as before with the exception of $k_1 = 10$.

Figure 3.8 clearly shows that for small times, the simulation produces a perfect match to the analytical solution. As time grows, so does the error especially close to the boundaries. The error values for this test case, calculated in the form of $L_2 = \sqrt{\frac{1}{N} \sum_{i=1}^{N} (T_{i}^{\text{SPH}} - T_{i}^{\text{exact}})^2}$, are plotted in figure 3.9.

**Comparison to Numerical Solution**

For numerical comparison, result generated by the ANSYS Fluent software (Canonsburg, PA, USA) is used as benchmark. A two-dimensional solid square plate with a dimensionless side length of 1 is considered. The transient heat conduction inside this square is captured as it is heated from its sides. For simplicity, the density, thermal conductivity, and heat capacity are all chosen to be unity as well. The plate is
Figure 3.8: Temperature profile across a slab with a temperature jump of 10 at $t = 0$, 1ms, 7ms, and 0.2s.

Figure 3.9: $L_2$ error values for temperature profile across a slab with a temperature jump of 10.
initially at zero temperature. At $t > 0$, the temperature of the walls is set to unity.

The test case is performed once using the SPH solver presented here, and once using
the Fluent solver. Transient temperature profile on the diagonal of the square and
the error values have been plotted in figures 3.10 and 3.11, respectively.

Figure 3.10: Temperature on the diagonal of the square for $t = 5, 25, 50, 75, 150,$
and $250ms$ (from bottom to top, respectively).

Figure 3.11: Difference in computed temperature values on the diagonal of the square
plate, $E = \max (|T_{SPH} - T_{Fluent}|)$. 
3.2.2 Two-dimensional Problem: Phase Change in the Corner of a Square

For the test cases described below, SPH particles are uniformly distributed with spacing of $\Delta x = \Delta y$ over the cross section of the cuboid as shown in figure 3.12. Domain is periodic in $z$ direction. Since each particle has a neighbourhood with a radius of $3\Delta x$, 3 layers of particles have been placed in the wall in order to make sure all inner particles have a complete neighbourhood. Depending how the values of gradients at wall particles are calculated, these 3 layers of particles might need to have a full neighbourhood of particles. Although test cases show results are not much affected by this, in order to have valid gradients calculated at these 3 layers of wall particles, an extra 3 more layers are also added in the walls here to make sure all wall particles that directly interact with inner particles have a full neighbourhood. The value of temperature of all wall particles are kept constant through all iterations.

![Figure 3.12: Initial positioning of fluid/solid particles (hollow squares) and wall particles (filled squares).](image)
Comparison to Numerical Results

The numerical results available in literature for the Stefan problem are used for numerical validation of the model in the presence of phase change. To simulate the Stefan problem, the three dimensional cuboid shown in figure 3.13 is filled with fluid which is at liquid state initially. Solidification will start as the temperature of the walls drop to a value below freezing. This problem has been widely used in literature as a benchmark. In most of reported cases, only a portion of cuboid cross section is simulated (1/4 or 1/8). The boundaries inside fluid for these cases are assumed adiabatic due to the symmetry in this problem. Here however the prism is fully modelled. In $z$ direction, a periodic boundary condition is imposed. For the wall, as SPH particles here see three particles immediate to them in their neighbourhood based on current configurations, three layers of particles with static $\rho H$ values are placed on the wall. In this way, it is made sure that all inner particles have complete wall neighbouring particles with wall temperature. All properties for solid and liquid are assumed to remain constant in each phase. Density is also assumed to be constant and equal for both phases.

Figure 3.13: Problem description: fluid is assumed to be inside a Cuboid which is infinitely long in $z$ direction. At the start of simulation, temperature at walls is suddenly dropped to a value below freezing temperature.

For the initial problem, fluid is assumed to be in liquid form and close to freezing
temperature. Wall temperature is suddenly dropped below freezing point and solidification starts from the boundaries toward the centre of cuboid. Fluid and solid properties are assumed to be constant and the values for non-dimensional parameters are $\theta = (T_i - T_w)/(T_m - T_w) = 1$ and $St = C_s(T_m - T_w)/L = 0.641$. Figure 3.14 shows the solidification front along the diagonal of cross section of the cuboid. For comparison, results reported by Cao et al. [52], Crowley [66], and Hsiao et al. [22] have also been plotted in this figure.

![Figure 3.14: Solidification front moving along the diagonal of a cross section of the cuboid for saturated liquid (compared against Cao et al. [52], Crowley [66], and Hsiao et al. [22]).](image)

The second problem has the same configuration as before with only the initial temperature being at a value larger than phase change temperature. Non-dimensional parameters for this problem are $\Theta = (T_i - T_w)/(T_m - T_w) = 9/7$ and $St = C_s(T_m - T_w)/L = 2$, $\alpha_l/\alpha_s = 0.9$, and $k_l/k_s = 0.9$. Figure 3.15 compares results for this test case against results of Cao et al. [52], Hsiao et al. [22], and Keung [67].
Effect of $\Delta T$

The same test case has also been conducted using the same properties and the assumption that phase change occurs at a temperature range of $\Delta T = 20$. As evident, results are not sensitive to allowing the phase change to occur over a temperature range.

Figure 3.15: Solidification front moving along the diagonal of a cross section of the cuboid for liquid above freezing temperature (Cao et al. [52], Hsiao et al. [22], and Keung [67]).

Effect of particle resolution

To make sure solution is not dependent to the initial particle positioning (particle resolution), different resolutions has been chosen for problems above. Figure 3.16 shows variation of results as resolution is changed from $10 \times 10$ to $100 \times 100$. It can be seen that by increasing the resolution, results converge to unique values. Results remain nearly identical for resolutions above these values.
Figure 3.16: Mesh resolution dependency. Figure shows solidification front moving along the diagonal of a cross section of a cuboid (which has a size of 2 by 2). Mesh resolution on the cross section has been changed from 10x10 to 100x100.

**Effect of Smoothing Kernel**

Effect of choosing different smoothing functions has also been studied by choosing four common smoothing functions shown in table 3.1. First example from before has been repeated using each of these kernels. Figure 3.17 shows that the choice of smoothing kernel will not affect final results.

### 3.2.3 Three-dimensional Problem: Droplet Impact & Solidification

The three-dimensional problem of interest here is impact and solidification of a molten drop. For comparison, test case conditions are chosen similar to test case of Aziz et al. [71] and Pasandideh et al. [72]. In this problem, a molten tin drop is impacted on a stainless steel substrate. The drop is initially at 240°C. The substrate is at room temperature. The impact is occurring at a low velocity of 1m/s. The drop has an initial diameter of 2.7mm. The spread factor \((D/D_0)\) for this test case has been
Figure 3.17: First example repeated using different kernels from table 3.1. Figure shows solidification front moving along the diagonal of a cross section of the cuboid for saturated liquid.

### Table 3.1: Smoothing kernels used in Enthalpy Formulation test cases

<table>
<thead>
<tr>
<th>Smoothing Kernels</th>
</tr>
</thead>
</table>
| **Meng et al. [68]**  
\[ W(R, h) = \frac{21}{16\pi h^3} \begin{cases} 
(1 - R/2)^4(2R + 1) & 0 \leq R \leq 2 \\
0 & \text{otherwise} 
\end{cases} \]  |
| **Johnson et al. [69]**  
\[ W(R, h) = \frac{5}{4\pi h^3} \begin{cases} 
(3/16)R^2 - (3/4)R + 3/4 & 0 \leq R \leq 2 \\
0 & \text{otherwise} 
\end{cases} \]  |
| **Monaghan et al. [70]**  
\[ W(R, h) = \frac{3}{2\pi h^3} \begin{cases} 
2/3 - R^2 + R^3/2 & 0 \leq R < 1 \\
(1/6)(2 - R)^3 & 1 \leq R < 2 \\
0 & \text{otherwise} 
\end{cases} \]  |
| **Liu et al. [44]**  
\[ W(R, h) = \frac{1}{4\pi h^3} \begin{cases} 
(2 - R)^3 - 4(1 - R)^3 & 0 \leq R \leq 1 \\
(2 - R)^3 & 1 < R \leq 2 \\
0 & \text{otherwise} 
\end{cases} \]  |
plotted in figure 3.18. Experimental results of Aziz et al. [71], and numerical result of Pasandideh et al. [72] have also been plotted here for comparison. Results indicate a good agreement to other numerical and experimental results for spread factor of the low velocity impacting drop. The final prediction for the spread factor here is closer to the numerical results of Pasandideh et al., both deviating near 6% from Aziz et al. experimental values.

![Spread factor comparison](image)

Figure 3.18: Spread factor ($D/D_0$) for a tin drop at 240°C with 2.7mm diameter impacting at 1m/s on a 25°C stainless steel substrate.

### 3.2.4 Mushy zone

Providing an exact benchmark for the mushy implementation methods seems impossible for various reasons. There are not any robust and accurate empirical measurements of the mushy region available to match the framework of this work. For the numerical studies that have been dedicated to this matter, the typical problem that has been studies is natural convection. In this case, the material is allowed to solidify in a cube (or corner in 2D). During this process, the natural convection resulting from temperature variations will accelerate molten liquid to flow similar to a lid-driven cav-
ity problem. This will result in an uneven growth of the solid front and can be used as a tool to measure the capability of mushy region handling. In the implementation in this work, however, the small time steps enforced on the simulation will make modeling of a natural convection cavity problem cumbersome. Therefore, instead a more relaxed problem has been chosen here to provide a qualitative comparison of the formulations. In this problem, solidification problem in a cavity under natural convection is studied. However, in order to avoid long wait times for the flow pattern to form, a divergence free velocity field in the form of \( v_\theta = 20 \frac{r}{R} \) and \( v_r = 0 \) is imposed on the fluid at the start of the simulation. Figure 3.19 shows schematic drawing of this problem. Liquid phase here is confined inside a cavity. The lower and upper walls are isolated. The fluid is allowed to have heat transfer only with a cold and hot source on each side. The red arrows in this figure show the initial velocity field in the domain.

Particle positions for this test case after the solidification has been partially completed is shown in figure 3.20 at \( t = 0.16 \). The solidification front (for \( \alpha = 0.5 \)) for equations 2.77 and 2.78 at this time has also been plotted in figure 3.21. It becomes evident that though both formulations used here provide similar results, the particle positions and arrangements are better preserved using 2.77. Hence, equation 2.77 has been used to capture mushy effects in the current study.
Figure 3.19: Divergence free initial velocity field for measuring effects of mushy region.
Figure 3.20: Particle positions in a $1 \times 1$ rectangle at $t = 0.16$ (s) colored by their liquid fractions. Mushy zone effected included using equation 2.77 (left) and equation 2.78 (right).
Figure 3.21: Solidification front (liquid fraction = 0.5) for equations 2.77 and 2.78 at $t = 0.16$. 
3.3 Phase-Change: Effective Heat Capacity

In the previous sections, inclusion of latent heat in energy equation with the help of enthalpy was reviewed. In the current section, procedures of adding latent heat to heat capacity is studied. Further details on these can be found at [73]

3.3.1 Two-Dimensional Problem: Phase Change in the Corner of a Square

The first test case is of solidification in the corner of a square initially filled with fluid, as shown in figure 3.22. The cuboid is assumed long enough in the z direction to form a 2D problem at its cross-section. For the solution here, a 3D setup of the domain has been used with a periodic boundary condition in the z direction. Solidification starts as the temperature of all side walls is suddenly changed to a value below the freezing point. From the analytical solution of this problem [74, 75], the solid-liquid interface that moves from the corner toward the square centre is of the form

\[ f(x^*) = [\lambda^m + C/(x^{*m} - \lambda^m)]^{1/m} \]  

(3.3)
where \( x^* = x/(4\alpha t)^{1/2} \), and \( \lambda \) is calculated by solving

\[
\frac{e^{-\lambda^2}}{\text{erf}\lambda} - \frac{T_i^* e^{-\lambda^2}}{\text{erfc}\lambda} = \sqrt{\pi} \beta \lambda
\]

(3.4)

where \( T_i^* = k_l/k_s(T_i - T_m)/(T_m - T_w) \) and \( \beta = 1/St \). Values of \( m \) and \( C \) depend on the non-dimensional parameters \( T_i \) and \( \beta \). For \( \beta = 0.25 \) and \( T_i^* = 0.3 \), these values are \( \lambda = 0.708 \), \( C = 0.159 \) and \( m = 5.02 \) \[74\]. The non-dimensional position of the solidification front on the cuboid diagonal (\( D \) in figure 3.22) can be also calculated from equation 3.3. On the diagonal, \( f(x^*) = x^* \). Solving equation 3.3 for this equality results in \( f(x^*) = 0.8958 \).

Numerically, SPH particles are uniformly distributed in all directions. Figure 3.23 shows the particle distribution at any \( x - y \) cross-section. Since each particle has a neighbourhood radius of \( 3\Delta x \), three layers of particles are placed in the wall so that all inner particles have a complete neighbourhood (filled circles in figure 3.23). The temperature of all wall particles is kept constant throughout the simulation. Properties of the solid and liquid are assumed to remain constant in each phase. Density is assumed to be the same in the two phases.

A comparison of numerical tests using each of the five equations versus the analytical solution is shown in figure 3.24. The error values are calculated by comparing the numerical solidification front position on the diagonal against the analytical value of \( 0.8958 \) using \( \left| \frac{x^* - 0.8958}{0.8958} \times 100 \right| \).

Results demonstrate that equations 2.65, 2.66, 2.72, and 2.73 yield nearly identical large errors, while the most accurate results are obtained using equation 2.76.
Comparison to Numerical Results

To compare the five equations against available numerical results, the solidification in the corner region is solved for the following non-dimensional parameters: $\theta = (T_i - T_w)/(T_m - T_w) = 9/7$, $St = C_s(T_m - T_w)/L = 2$, $\alpha_l/\alpha_s = 0.9$, and $k_l/k_s = 0.9$. The tests conducted here have a fine particle resolution of $100 \times 100$ particles in the $x - y$ cross-section. Results for this case are plotted in figure 3.25. Numerical results of the same problem from Cao et al. [52], Hsiao et al. [22], and Keung [67] using a coarser mesh of $20 \times 20$ are also shown. All methods converge to the same solution.

Effect of particle resolution

To assess mesh dependency, the previous problem is solved on a much coarser $30 \times 30$ mesh. Results are shown in figure 3.26 along with the same results of Hsiao et al. [22], Cao et al. [52], and Keung [67] with a mesh resolution of $20 \times 20$. It is clear that by reducing the resolution, equations 2.72 and 2.73 start to deviate from the converged
values, while equations 2.65, 2.66, and 2.76 show near to no sensitivity to this decrease in solution resolution. This shows how these equations are more accurate at coarser resolutions.

As figure 3.24 shows that equation 2.76 is the most accurate, the rest of this section is focused on further examination of the functionality of this equation.

Figure 3.27 illustrates the melting of a cuboid, for $\theta = 0.3, St = 4, \alpha_l/\alpha_s = 1,$ and $k_l/k_s = 1$. Results are compared at two resolutions, $40 \times 40$ and $100 \times 100$, at $0.1, 0.4, 0.8, 1.2$, and $1.44$ seconds (figure 3.27). The solid lines in this figure show the analytical solution.

Effect of $\Delta T$

Equation 2.76 assumes that phase change occurs over a temperature range $\Delta T$, the value of which must be chosen for each problem. It was mentioned previously that the value of $\Delta T$ must be small enough to approximate physical reality, yet if $\Delta T$ is too small, some particles might not experience phase change as the temperature can
jump from a value above (below) melting to a value below (above) in one time step during solidification (melting). This must be taken into consideration when choosing $\Delta T$ and $\Delta t$. For the previous test cases, a dimensionless value of $\Delta T = 0.02$ was used.

The melting cuboid problem is considered again, with $T_i^* = 0.3$, $St = 4$, $\alpha_l/\alpha_s = 1$, and $k_l/k_s = 1$. $\Delta T$ is set to values of 0.00001, 0.01, 0.02, 0.05, and 0.08. The error values for this test case, compared to the analytical solution, are plotted in figure 3.28. For very small $\Delta T$ (0.00001 here), errors are large as many particles do not experience the latent heat, and at large $\Delta T$ (0.08 here), the error grows as the phase change temperature interval deviates too far from the physical reality. The results of various test cases, including the one reported here, confirm that there exists a large range of $\Delta T$ over which the results are relatively independent of the choice of $\Delta T$. 

Figure 3.25: Position of the solidification front on the diagonal cross section of the cuboid (resolution: $100 \times 100$)
Finally, the choice of smoothing kernel that appears in equation 2.75 is important as it dictates how the latent heat is released into the domain. Table 3.2 lists three smoothing functions (from [68–70]). These kernels are plotted in figure 3.29. To compare these kernels, $W_T$ and $W_P$ are taken to be identical, where the former is in the 1D domain of temperature and the latter is in the 3D $x-y-z$ domain. This only changes the constant coefficients of these kernels. For instance, the Meng [68] kernel in 3D has a coefficient of $21/(16\pi h^3)$ for $W_P$ and a coefficient of $3/4h$ (in 1D) for $W_T$. Note that the $\alpha^{3D}$ values are used for the 2D test case here, since it is solved in a 3D domain.

For the case of the solidification of the cuboid, results using these three kernels are compared to the analytical solution. Figure 3.30 shows that all three kernels produce accurate results with an error below 5%, but the Meng et al. [68] and Johnson et al. [69] kernels produce somewhat more accurate solutions.
Table 3.2: Smoothing kernels

<table>
<thead>
<tr>
<th>Kernel</th>
<th>$\alpha^{3D}$</th>
<th>$\alpha^{1D}$</th>
<th>Ref</th>
</tr>
</thead>
<tbody>
<tr>
<td>$W(R, h) = \alpha \begin{cases} (1 - R/2)^4(2R + 1) &amp; 0 \leq R \leq 2 \ 0 &amp; otherwise \end{cases}$</td>
<td>$\frac{21}{16\pi h^3}$</td>
<td>$\frac{3}{4h}$</td>
<td>[68]</td>
</tr>
<tr>
<td>$W(R, h) = \alpha \begin{cases} (3/16)R^2 - (3/4)R + 3/4 &amp; 0 \leq R \leq 2 \ 0 &amp; otherwise \end{cases}$</td>
<td>$\frac{5}{4\pi h^3}$</td>
<td>$\frac{1}{h}$</td>
<td>[69]</td>
</tr>
<tr>
<td>$W(R, h) = \alpha \begin{cases} 2/3 - R^2 + R^3/2 &amp; 0 \leq R &lt; 1 \ (1/6)(2 - R)^3 &amp; 1 \leq R &lt; 2 \ 0 &amp; otherwise \end{cases}$</td>
<td>$\frac{3}{2\pi h^3}$</td>
<td>$\frac{1}{h}$</td>
<td>[70]</td>
</tr>
</tbody>
</table>

### 3.3.2 Three-Dimensional Problems

In this section, the validity of the model of equation 2.76 is assessed on two 3D test cases.

#### Melting of a sphere

The first 3D test case is the melting of a sphere. Problem specifications are the same as in [76]. A sphere is considered to be initially at its melting temperature ($T_i = 0^\circ C$). At time 0, the temperature of the wall surrounding the sphere is raised to a value higher than the melting temperature ($T_w = 1^\circ C$). In this test case, similar to the test performed by [76], the Stefan number $St$ is varied from 1 to 4. The sphere has a radius of 0.25m and specific heats and thermal diffusivities are set to unity. Figure 3.31 shows the total melting time for the sphere at different Stefan numbers. The melting times obtained using equation 2.76, as can be seen in figure 3.31, are in good agreement with the mathematical model of [77].

Particle temperatures for the case of $St = 4$ are plotted in figure 3.32 at $t = 0$, 0.0025, and 0.0065.
Melting of a cone

The second 3D test is the melting of a cone. For comparison, the problem specifications have been chosen similar to a test case from [78]. A cone with a half angle of 30 degrees is initially at $T_i = 0^\circ C$. The temperature on the wall surrounding the cone is increased to $T_w = 0.3^\circ C$ at $t = 0$. The phase change interface at different times is shown in figure 3.33, and compared to the results of Ayasoufi et al. [78] (CE/SE method). As can be seen, the SPH solution yields results close to those of [78], while slightly underpredicting the melt front.

3.3.3 Computational Cost

Finally, the SPH solver used here was developed to run entirely on graphics processing units (GPU). The device used for this study is the NVIDIA® Tesla® K20 GPU accelerator with 2496 processor cores and a processor core clock of 706 MHz. For the sample conduction problems with phase change, 10.3 kB of memory per particle was needed on the GPU device. The five equations mentioned before perform quite consistently with regard to the amount of computational time, except for equation 2.72, which is near 8% faster than the rest. This is because equation 2.72 approximates the amount of latent heat released (absorbed) without the need of smoothing steps or evaluating gradients. However, equation 2.72 is also the least accurate. Of the rest of the equations, equation 2.76 requires the least number of computation steps, and so is easier to implement.
Figure 3.27: Melting of a cuboid at 0.1, 0.4, 0.8, 1.2, and 1.44 seconds. Solid lines are the analytical solution. Solution resolution: $40 \times 40$ (top) and $100 \times 100$ (bottom).
Figure 3.28: Error of the solidification front position versus the analytical solution, for solidification in a corner, for various $\Delta T$.

Figure 3.29: $W(R, h)/h^3$ for the three smoothing kernels.
Figure 3.30: Error of the solidification front position versus the analytical solution, for three different kernels.

Figure 3.31: Melting of a sphere. Total time of melting versus Stefan number. Squares show results obtained using the model presented here, and the solid line is from the mathematical model of [77].
Figure 3.32: Melting of a sphere of radius 0.25, with a wall temperature $T_w = 1^\circ C$, at $t = 0, 0.0025$, and 0.0065 seconds. Left: un-melted particles; Right: temperature.
Figure 3.33: Melting of a cone. Interface position is plotted at different times: 0.05, 0.1, 0.25, and 0.5. Red crosses are the results of Ayasoufi et al. [78] obtained using a space-time conservation element and solution element (CE/SE) method.
Chapter 3. Validation

3.4 Choice of the Right Model

The introduction of the two main models presented here for the implementation of latent heat, i.e. adding it as source term in enthalpy formulation or using the modified effective heat capacity, raises the question of which model should be used for any particular problem. Under the test conditions here, the two methods have performed in a very similar manner. For instance, the results of the solidification in a corner conducted for both models shown in figures 3.14 and 3.25 have been combined here in figure 3.34. There are however some advantages and disadvantages to each model that might make them more suitable for different applications.

![Figure 3.34: Position of the solidification front on the diagonal cross section of the cuboid - comparing equations 2.59, 2.76, and numerical results of Cao [52].](image)

The inclusion of latent heat in an enthalpy formulation (equation 2.59) for instance has the advantage of modeling phase change for pure materials without the need of a phase change temperature interval. This can be very helpful, especially since the phase change interval for pure materials is completely artificial and any non-zero value will lead to introducing some inevitable amount of error (which of course can be limited if the interval is chosen small enough).
Comparing the performance of each of these models with the choice of different smoothing kernels shown in figures 3.15 and 3.29 reveals that the enthalpy formulation is less sensitive to the smoothing function. This comes from the fact that the effective heat capacity method relies heavily on the smoothing kernels to determine the amount of latent heat that is released at each particle. The enthalpy model however does not have that reliance and hence becomes a more suitable candidate for free surface problems where the neighborhood of smoothing kernels become interrupted.

The effective heat capacity method, however, demands less computations compared to the enthalpy formulation. Implementation of this method is also straightforward and the calculations can be performed in less computational steps. Effective heat capacity model, can be also applied to convective problems by making the assumption that compressibility effects remain negligible in calculations. This is since the term making the difference between convective and non-convective energy equations in Lagrangian form is directly proportional to density variations, and hence vanishes for the case of complete incompressibility.
Chapter 4

Suspension Plasma Spraying (SPS)

In the following chapter, a comprehensive numerical study on SPS process is performed. Effects of different parameters, such as torch operating condition and injection angle, on the final formation of molten ceramic droplets are studied. Results obtained in this chapter will form the ground work for next chapter, where impact and solidification of molten SPS droplets are studied. Further details of the SPS simulations can be found at [79,80].

4.1 Problem Specification

A schematic drawing of the plasma torch and suspension injection used in the current study is shown in 4.1. The plasma torch used in current study has a nozzle exit diameter of 6mm and an anode length of 20mm (3MB plasma spray gun, Orlikon-Metco, Switzerland). The operating conditions used for this torch vary for different test cases and have been summarized in table 4.1.

Thermodynamic and physical properties for materials have also been shown in table 4.2 [81–83]. Suspension material of interest here is a mixture of Yttria-stabilized...
zirconia (10\% wt.) and water. The suspension droplets are injected in the domain at a uniform diameter of 150\textmu m. For this study, dispersion of the solid content is neglected and instead, it is assumed that the solid content forms a concentrated ball inside the suspension droplets. Injection site is varied using the axial and radial distances of the injector from the nozzle exit. Suspension particles can also be injected with different initial velocity vectors. For the test cases where the particles are collected on a flat substrate, the standoff distance is 80mm.

### 4.2 Numerical Methodology

Different numerical methods and treatments have been used in the current study. The Finite Volume based ANSYS Fluent V14.5 (Canonsburg, PA, USA) has been used as the main solver. User defined functions (UDF) written in C programming language
Table 4.1: Summary of operating conditions

<table>
<thead>
<tr>
<th>Torch geometry and operating conditions</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Exit nozzle diameter (mm)</td>
<td>6</td>
</tr>
<tr>
<td>Anode length (mm)</td>
<td>20</td>
</tr>
<tr>
<td>Current (A)</td>
<td>450 - 600</td>
</tr>
<tr>
<td>Voltage (V)</td>
<td>24.8 - 76.8</td>
</tr>
<tr>
<td>Thermal efficiency (%)</td>
<td>47.1 - 51.2</td>
</tr>
<tr>
<td>Ar/H2 mass flow inlet (slpm)</td>
<td>35.4 - 140</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Suspension specifications</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Ceramic content</td>
<td>Yttria-stabilized zirconia (YSZ)</td>
</tr>
<tr>
<td>Base fluid</td>
<td>Water</td>
</tr>
<tr>
<td>Suspension droplet diameter (µm)</td>
<td>150</td>
</tr>
<tr>
<td>Solid fraction (%wt, %vol)</td>
<td>10, 1.96</td>
</tr>
</tbody>
</table>

Table 4.2: Materials properties

<table>
<thead>
<tr>
<th>YSZ</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>ρ (kg/m³)</td>
<td>5560</td>
</tr>
<tr>
<td>µ (Pa·s)</td>
<td>0.029</td>
</tr>
<tr>
<td>σ (N/m)</td>
<td>0.43</td>
</tr>
<tr>
<td>Cₚ (J/(kg·K))</td>
<td>678.5, T &gt; 873</td>
</tr>
<tr>
<td>ΔHsf (kJ/kg)</td>
<td>710</td>
</tr>
<tr>
<td>k (W/(m·K))</td>
<td>2.4</td>
</tr>
<tr>
<td>Tₘₚ (K)</td>
<td>2975</td>
</tr>
<tr>
<td>ΔHfg (kJ/mol)</td>
<td>Equation 4.21</td>
</tr>
<tr>
<td>k (W/(m·K))</td>
<td>0.6</td>
</tr>
<tr>
<td>Tbp (K)</td>
<td>373</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Water</th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>ρ (kg/m³)</td>
<td>998.2</td>
</tr>
<tr>
<td>µ (Pa·s)</td>
<td>0.001003</td>
</tr>
<tr>
<td>σ (N/m)</td>
<td>0.038 (YSZ-Water mixture)</td>
</tr>
<tr>
<td>Cₚ (J/(kg·K))</td>
<td>4182</td>
</tr>
<tr>
<td>ΔHfg (kJ/mol)</td>
<td>Equation 4.21</td>
</tr>
<tr>
<td>k (W/(m·K))</td>
<td>0.6</td>
</tr>
<tr>
<td>Tbp (K)</td>
<td>373</td>
</tr>
</tbody>
</table>

are then used for implementing procedures not originally supported by the Fluent solver. The simulation process can be broken into two main stages. Initially, the flow of the plasma gas exiting the nozzle at high speed and temperature is calculated.
The discrete phase models are then used to track suspension droplets inside the flow. To avoid numerical complications, instead of injecting a continuous stream of suspension at the beginning, a train of already atomized suspension droplets is injected into the flow. The momentum and heat transfer equations for the gas and particles are coupled in order to capture the effect of particles on the gas flow and vice versa. The evaporation of the liquid phase (here water) inside suspension droplets is reflected in the gas flow by adding species source terms for water vapour. In the following sections, governing equations used in the numerical simulation of the torch and suspension droplets are reviewed.

### 4.2.1 Torch Model

Detailed modelling of the plasma torch, involving phenomena like flow of gas in the nozzle, chemical reactions, and generation and fluctuations of the arc can be found in studies like [84–88]. Here however a simpler approach is chosen. The plasma torch model is schematically shown in Fig. 4.1. The plume is simplified to a jet of fluid entering the domain at high speed and temperature. The torch carries a premixed Ar-$H_2$ gas mixture into atmospheric air. As suspension droplets get injected into the torch, evaporation of liquid phase will also add water vapour to the plasma gas composition. At each cell in the domain, temperature-dependant thermodynamic properties of the plasma gas are calculated based on the mass/volume fraction of each component in this mixture. The chemical reactions and arc influences are neglected. Instead, a volumetric heat source is added inside the torch nozzle. Using torch operating conditions, i.e., current ($I$), voltage ($V$), and thermal efficiency ($\eta$), this volumetric heat source can be estimated as

$$Q = \eta EI/\Omega$$  \hspace{1cm} (4.1)
Ω is the volume of anode. Inclusion of thermal efficiency eliminates the need for adding heat losses at the torch walls and radiation losses. In order to avoid a long torch entrance length, a fully developed turbulent velocity profile in the form of power-law is taken at the Ar-H₂ inlet, i.e.,

\[ u(r) = u_{max} \left( \frac{r}{R} \right)^{1/n} \]  

(4.2)

For the turbulence model, different models and resolutions were tested here. The Realizable k-ε model with the standard wall functions was found to produce the best results at a low computational cost. To avoid deficiencies of the k-ε model in axisymmetric frameworks [89], the flow domain has been fully resolved in three dimensions here. Although this eventually adds to the computation time, it was proven to be useful in better capturing trajectories of the suspension droplets injected into the flow in 3D. The extra computational cost is also partially recovered as the k-ε model is computationally less expensive compared to other methods such as RSM [82]. The turbulent intensity at the inlet is approximately [82]

\[ I_t = 0.16 Re_{DH}^{-1/8} \]  

(4.3)

\( Re_{DH} \) is the Reynolds number based on the hydraulic diameter of the torch nozzle. Using this prescribed turbulent intensity, the turbulent kinetic energy is calculated as

\[ k = 3/2 u_{avg}^2 I_t^2 \]  

(4.4)

and the value of turbulent dissipation rate is evaluated using

\[ \epsilon = C_{\mu}^{3/4} k^{3/2} \]  

(4.5)
\( C_\mu = 0.09 \) is an empirical constant, and \( L \) is the length scale approximated to be 0.07 of the hydraulic diameter.

### 4.2.2 Suspension Particle Model

To avoid numerical complications of primary fragmentation of liquid jets, separate droplets are injected rather than a continuous jet of suspension liquid. The droplets are tracked in a Lagrangian framework as they travel in the domain. The liquid phase in the suspension starts evaporating as the droplets receive heat from the plasma flow. Heat losses due to radiation from the droplets to the surroundings are also included. The plasma plume however is assumed to be optically thin. As the liquid phase vanishes, the concentration of solid contents in the suspension grows. This leads to change in some physical properties, especially viscosity. When the solvent has completely evaporated, the solid contents are tracked until they reach the melting point. After melting, physical properties of particles is the same as that of the molten ceramic. In the following sections, equations and models governing each of these transitions are presented in detail.

### 4.2.3 Momentum

Suspension droplets/particles here are tracked in Lagrangian manner using a two-way coupled discrete phase model. Plasma gas with the velocity \( u \) accelerates particles inside the domain. Particle acceleration is calculated using a balance between forces acting on the particles and their inertia in the form of [82]

\[
\frac{du_p}{dt} = F_D (u - u_p) + F \tag{4.6}
\]
Here, $F_D$ is the drag force given as

$$F_D = 18\mu \rho_p d_p^2 C_D Re \frac{C_D}{24}$$

(4.7)

The drag coefficient $C_D$ is calculated from instantaneous Reynolds number of each particle

$$C_{Df} = \frac{24}{Re}$$

(4.8)

for $Re < 0.01$,

$$C_{Df} = \frac{24}{Re} + 3.156 Re^{-0.18 + 0.05 \log Re}$$

(4.9)

for $0.01 < Re < 0.2$, and

$$C_{Df} = \frac{24}{Re} + \frac{6}{1 + \sqrt{Re}} + 0.4$$

(4.10)

for the range $0.2 < Re < 10^5$ based on empirical correlations for small spherical particle sizes $(5 - 100\mu m)$ [90]. For particles moving in plasma flow, this drag coefficient needs to be modified to take effects of variable properties due to temperature gradient and non-continuum (Knudsen effects) into account. Influence of Basset history term is neglected for particles smaller than $100\mu m$ [91]. Effect of variable property is added using proposed method of Lee et al. [92]. Non-continuum effects also become important for Knudsen number regime $10^{-2} < Kn < 1$ [91]. This is also considered using correction terms of Chyou et al. [93] and Chen et al. [94]. A superposition of these corrections, as proposed by Pfender [91], can be used to calculate the final drag coefficient in the form of

$$C_D = C_{Df} f_1 f_2$$

(4.11)

where correction factors for strong variation of properties and non-continuum effects
are

\[ f_1 = \frac{\rho_\infty \mu_\infty}{\rho_f \mu_f}^{-0.45} \]

\[ f_2 = \left[ 1 + \frac{2 - a}{a} \frac{\gamma}{1 + \gamma} \frac{4}{Pr_f} Kn^* \right]^{-0.45} \]  \hspace{1cm} (4.12)

The subscript \( f \) means properties are calculated at particle’s film temperature, \((T_{\text{cell}} + T_p)/2\). \( a \) is the thermal accommodation coefficient, \( \gamma \) is the heat capacity ratio, and \( Pr_f \) is the gas Prandtl number evaluated at film temperature. Here, \( Kn^* \) is the Knudsen number based on using effective molecular mean free path (\( \lambda_{eff} \)) and particle diameter (\( d_p \)). \( \lambda_{eff} \) is calculated from

\[ \lambda_{eff} = \frac{2K}{\rho_f V_f C_p Pr_f} \]  \hspace{1cm} (4.13)

with \( V_f, C_p, \) and \( K \) being average thermal velocity, specific heat and thermal conductivity, respectively. \( F \) in equation 4.6 takes thermophoretic effects into account. Thermophoretic forces are exerted on small particles travelling in gas with temperature gradient and are in the opposite direction to this gradient [95]. This force is added using proposed method of Talbot et al. [96]

\[ F = -\frac{6\pi d_p \mu^2 C_s (K^* + C_t Kn)}{\rho (1 + 3C_m Kn) (1 + 2K^* + 2C_t Kn)} \frac{1}{m_p T} \frac{\partial T}{\partial x} \]  \hspace{1cm} (4.14)

\( K^* \) is the ratio of fluid to particle thermal conductivities, \( m_p \) is the mass of particle, \( C_s = 1.17, C_t = 2.18, \) and \( C_m = 1.14 \). Brownian Forces are neglected since there are not many sub-micron particles involved in test cases here. The effects of turbulence on dispersing the particles is included using stochastic tracking method. This method calculates particle trajectories taking instantaneous turbulent velocity fluctuations into account.
4.2.4 Breakup model

The suspension jet after injection will experience several stages of breakup before reaching the substrate. These breakups need to be included in the numerical method. Melliot et al. [97] numerically studied breakup of a train of YSZ-water suspension droplets in the plasma gas. Similarly, Vincent et al. [98] used LES-VOF to capture the atomization process of a continuous liquid jet of water injected into the plasma flow. In these simulations, evaporation of liquid phase is neglected. Moreover, the domain of study has been limited to the area close to nozzle exit. Implementation of numerical simulations like these into largescale models, like the one used in the current study, makes solution computationally expensive. Direct simulation of atomization process demands a very fine mesh resolution at fragmentation points and also solving a fluid surface tracking model like VOF [99]. To overcome these issues in the current study, suspension is injected into the domain in the form of droplets. This means that the primary breakup has already taken place. For the secondary breakups and the rest, KHRT (Kelvin-Helmholtz/Rayleigh-Taylor) model [27, 28] is used. This method, which is applicable to high Weber number flows [82], combines effect of aerodynamic forces which create Kelvin-Helmholtz waves along with Rayleigh-Taylor instabilities caused by acceleration of drops into the free stream. These mechanisms capture breakup using drop’s surface wave growth: fastest growing instability will cause the drop to breakup. For the suspension of study here, the viscosity is updated at each iteration based on drop’s concentration. Breakup model is also influenced by this change of viscosity. Aggregate explosions are neglected here, hence no breakup occurs till all the solvent is evaporated and all solid content has melted. When a molten drop is formed from solid material, breakup is resumed.
4.2.5 Heat transfer

Particle heating and cooling due to convective heat transfer, evaporation, and radiation at particle’s surface is governed by [95]

\[ m_p C_p \frac{dT_p}{dt} = h A_p (T_\infty - T_p) + \epsilon A_p \sigma (T_\infty^4 - T_p^4) - \frac{dm_p}{dt} \Delta H_{fg} \] (4.15)

\( \sigma \) is the Stephan Boltzmann constant, \( \epsilon \) is the emissivity for particles, \( m_p \) is the mass of each particle, \( C_p \) is the heat capacity, \( T_\infty \) is the ambient temperature, \( T_p \) is the particle temperature, \( h \) is the convective heat transfer coefficient, \( dm_p/dt \) is the evaporation rate, and \( h_{fg} \) is the latent heat of the liquid phase. The convective heat transfer coefficient is calculated with the help of Nusselt number using Ranz-Marshall correlation

\[ Nu_f = 2.0 + 0.6 Re_d^{1/2} Pr^{1/3} \] (4.16)

\( Re_d \) is Reynolds number based on particle diameter and \( Pr \) is gas phase Prandtl number. Similar to momentum, effects of temperature gradients and Knudsen number are included using correction factors. Final Nusselt number is corrected using [94,100]

\[ Nu = \frac{h d_p}{k_\infty} = Nu_f f_{h1} f_{h2} \] (4.17)

with \( f_{h1} f_{h2} = (f_1 f_2)^{\frac{\pi}{6 \nu}} \). As the solvent inside particles evaporates, heat transfer mechanisms changes. If initial suspension has low ceramic concentration, a shell of molten ceramic might form imprisoning solvent inside [39]. Also during solvent evaporation, the cloud of ceramic particles may get scattered as a result of sudden explosion. Taking all these effects into account is nearly impossible due to lack of sufficient empirical correlations. Hence, it is reasonable to make simplifying assumptions. Inclusion of explosion and shell formation effects are left for future studies. Instead,
it is assumed that evaporation continues till all the liquid content in the droplets is evaporated. Prohibiting scattering of solid particles before they are melted together will possibly result in a larger predictions for final particle sizes.

### 4.2.6 Evaporation of the liquid phase

During initial stages of droplets’ flight, the liquid phase starts to evaporate. As this phase evaporates, the suspension’s concentration varies significantly as it goes from being dilute to becoming a slurry of moist solid particles that will dry out. The evaporation rate can be corrected to match the drying process of solid-liquid mixtures. Evaporation of the solid content is negligible through all stages. The drying rate of the mixture can be calculated using two different methods. Where experimental data for calibration is available, the model presented by Stendal [101] and Seader et al. [102] for solid-liquid slurries can be used to estimate the evaporation rate

\[
\frac{dm_p}{dt} = m_p \frac{dW}{dt} = m_p f(W^*) \exp \left( - \frac{\Delta H_{fg}}{RT_p} \right)
\]

\[W^*\] is the wet basis moisture content, and \(R\) the universal gas constant. \(f(W^*)\) is evaluated using experimental data for every particular suspension. Where empirical data for calculation of \(f(W^*)\) is not available, the evaporation and mass transfer rates can be obtained using Sherwood number correlation [95]

\[
Sh = \frac{k_c d_p}{D} = 2 + 0.6 Re_{DH}^{0.5} Sc^{1/3}
\]

\(D\) is the binary diffusion coefficient, Schmidt number is \(Sc = \mu/\rho D\), and \(k_c\) is the mass transfer coefficient. By taking an estimate for the binary diffusion coefficient, the mass transfer coefficient becomes available using equation 4.19. Therefore, the
evaporation rate can be calculated using

$$\frac{dm_p}{dt} = k_c M A_p (C_s - C_b)$$ (4.20)

where $M$ is the molecular weight of the liquid content, and $C_s$ and $C_b$ are vapour concentrations at drop’s surface and the bulk of gas, respectively. The latent heat of evaporation here is evaluated at particle temperature using

$$\Delta h_{fg} = -0.0438926 T_p + 57.0735 (kJ/mol)$$ (4.21)

for water [103] and using

$$\Delta h_{fg} = A \exp \left( -\alpha \frac{T_p}{T_c} \right) \left( 1 - \frac{T_p}{T_c} \right)^\beta (kJ/mol)$$ (4.22)

for ethanol from [104, 105], with $A=50.43$ kJ/mol, $T_c=513.9$ K, $\alpha=-0.4475$, and $\beta=0.4989$. As particles go through multiple breakups before the liquid phase is completely evaporated, the solid content of each particle is estimated using the assumption that each breakup divides the solid content in the particle evenly between the forming children based on their diameters.

### 4.2.7 Melting of the solid content

After the liquid phase is completely evaporated, the solid content is heated till reaching the melting point of the solid material. $C_p$ values for particles are now only calculated from the properties of the solid material. Viscosity values are also adjusted in the absence of liquid phase. This means that when the fraction of liquid content in each drop becomes negligible, the viscosity for the mixture, which now
consists of only solid particles, is set to a large value to mimic non-molten solid matter. This is demonstrated in Fig. 4.2 for YSZ. In a 10K window around the melting temperature, the viscosity is linearly dropped from this large value to the value for molten YSZ. This will also be an estimate for the mushy behaviour of the melting process.

![Figure 4.2: Mushy zone effect on viscosity values of YSZ.](image)

The latent heat consumed during melting process is also included by modifying $C_p$ values of YSZ. This is done using

$$C = \begin{cases} C_s, & \text{if } T \leq T_1 \\ C_m + L/(T_2 - T_1), & \text{if } T_1 < T < T_2 \\ C_l, & \text{if } T_2 \leq T \end{cases}$$

(4.23)

Where $T_1$ and $T_2$ mark the temperature range over which the melting occurs, and $L$, is the latent heat. For simplicity, $C_m$ is chosen to be $(C_s + C_l)/2$ here. This also demonstrated in figure 4.3 for YSZ suspension.
Surface tension coefficient also undergoes various changes during the evaporation of the liquid phase. As a wide-ranging set of information on surface tension versus concentration is not available for ceramic suspensions, in the current study, when liquid phase is present in droplets, surface tension is approximated to have the same value as that of the liquid phase. When the liquid phase is evaporated and the solid content has melted, the surface tension of the molten ceramic is used thereafter. In between these two limits, no breakup of the droplets is allowed and hence the surface tension values are not of interest.

4.2.8 Effective viscosity

With the evaporation of the liquid phase, the concentration of solid matter in droplets changes. Consequently, the viscosity of the mixture needs to be updated. Many studies have used analytical and empirical tools for evaluating suspension viscosities at different solid content concentrations. Einstein’s viscosity model is among the first
that predicts viscosity of a dilute suspension of spherical droplets using [106]

$$\mu_r = 1 + 2.5\phi$$  \hspace{1cm} (4.24)

with $\phi$ being the volume fraction of solid particles in suspension ($V_{\text{solid}}/V_{\text{total}}$). For higher concentrations, this equation was modified by Guth et al. [107] to take interaction between solid particles into account in the form of

$$\mu_r = 1 + 2.5\phi + 14.1\phi^2$$  \hspace{1cm} (4.25)

where $\mu_r$ is the relative viscosity of the suspension to the pure fluid viscosity, shown as

$$\mu_r = \frac{\mu_{\text{eff}}}{\mu_{\text{liq}}}$$  \hspace{1cm} (4.26)

Using empirical data, Thomas enhanced this model to form [108]

$$\mu_r = 1 + 2.5\phi + 10.05\phi^2 + A\phi^{B\phi}$$  \hspace{1cm} (4.27)

where $A = 0.00273$ and $B = 16.6$. For larger particles at higher concentrations and by taking effects of particle interactions into account, Toda el al. [38] proposed using

$$\mu_r = \frac{1 + 0.5k\phi - \phi}{(1 - k\phi)^2(1 - \phi)}$$  \hspace{1cm} (4.28)

$k$ is a parameter obtained using empirical results and has been reported to be in the form of $k = 1 + 0.6\phi$ for spherical particles in water (5$\mu$m radius). Models mentioned above do not take effects of maximum packing fraction, $\mu_m$, into account. $\phi_m$ is the largest possible volume fraction that can be achieved by adding solid particles to the suspension. Physical shape of particles along with their electric charge can affect the
value of $\phi_m$. Since theoretically suspension viscosity should converge to infinity for solid concentrations close to $\phi_m$ [40], it is important to include this parameter into the model. Krieger et al. [109] have proposed inclusion of $\phi_m$ in the form of

$$\mu_r = 1 - \frac{\phi}{\phi_m}^{-[\mu]_m}$$  \hspace{1cm} (4.29)

Here $[\mu]$ is the intrinsic viscosity defined as the limiting value, which dominates suspension behaviour at low concentrations [110] and is given by [111]

$$[\mu] = \lim_{\phi\to 0} \lim_{\dot{\gamma}\to 0} \left( \frac{\mu_r - 1}{\phi} \right)$$  \hspace{1cm} (4.30)

It is common to approximate $[\mu]$ to 2.5, which is derived by applying equation 4.30 to Einstein’s model in equation 4.24. Dabak et al. [112] also proposed a model containing another empirically adjustable variable, $N$, given as

$$\mu_r = \left[ 1 + \frac{[\mu] \phi}{N \left( 1 - \frac{\phi}{\phi_m} \right)} \right]^N$$  \hspace{1cm} (4.31)

$N$ is a flow related variable and is taken to be 2 for high shear rates [113]. More recently, Senapati et al. [114] has proposed an improved model that takes effects of particles size distribution along with shear rate into account

$$\mu_r = S \left[ 1 + \frac{[\mu] \phi}{\dot{\gamma}^n \phi_m - \phi} \right]^N$$  \hspace{1cm} (4.32)

where $\dot{\gamma}$ is the shear rate, and $n$ is the flow behavior index in the power law equation ($\tau = K \dot{\gamma}^n$). Parameter $S$ is calculated from $S = 10C_U d_{50}$ where $C_U$ is the coefficient of uniformity calculated from $C_U = d_{60} d_{10}$. Predictions of models mentioned above
show deficiencies when were tested here against available data for ceramic suspensions. A more recent model by Horri et al. [115] however shows close predictions for ceramic suspensions. The relative viscosity of Horri, which is valid over a wide range of concentrations and shear rates, is in the form of

$$\mu_r = 1 + 2.5\phi + K\phi \left(\frac{\phi}{\phi_m - \phi}\right)^2$$  \hspace{1cm} (4.33)

Parameter $K$ is determined using experimental data and varies for different shear rates. For 8 mol% YSZ-water suspension using experimental results of Arevalo-Quintero et al. [116] (as plotted in figure 4.4, $\rho_s=5.54\text{g (cm}^3\text{)}$, $\rho_l=0.997\text{ g (cm}^3\text{)}$, $\mu_l=0.89\text{mPas}$), under a constant shear rate with $\phi_m=21.76$, parameter $K$ can be estimated to be $K = 5.660117$. As equation 4.33 produces good fits to experimental measurements of ceramic suspensions of interest here, it has been used in this study for prediction of ceramic viscosities.

![Figure 4.4: Viscosity of YSZ-Water suspension under different concentrations of YSZ powder (from Arevalo-Quintero et al. [116]). Dashed line is the trend line.](image)

$$\mu_{eff} = 0.0035\phi^3 - 0.0501\phi^2 + 0.2651\phi + 0.7279$$

$R^2 = 0.9994$
4.2.9 Maximum packing fraction ($\phi_m$)

Most correlations used for viscosity approximation rely heavily on a good approximation of the maximum packing fraction. Two different methods of finding $\phi_m$ are reviewed here. The first method will be employed when experimental results for suspension viscosity at high concentrations is available. Where experimental data is missing, analytical models that are described below can be used instead.

4.2.10 $\phi_m$ calculation from empirical data

For the case where experimental data for suspension viscosity is available, the method explained by Senapati et al. [114] can be used to approximate $\phi_m$. Experimental results of Arevalo-Quintero et al. [116] for viscosity of YSZ suspension in different solid contents (figure 4.4) is used here as an example. The suspension used in their experiment was prepared using water and 8 mol% YSZ powder with $d_{10}$, $d_{50}$, and $d_{90}$ values close to $0.8\mu m$, $2.6\mu m$, and $6.3\mu m$, respectively. The limiting value of $1/\mu_r$ when it goes to zero corresponds to $\phi_m$. Using values presented in figure 4.4 for viscosity of YSZ suspensions, $1/\mu_r$ can be calculated as shown in figure 4.5. Using a trend line, or trial and error, the place for which $1/\mu_r$ has a very small value can be found here as $\phi_m = 21.76$.

Parameter $N$, appearing in equations 4.31 and 4.32, is also a suspension dependant parameter and needs to be calculated from experimental data. As indicated by Senapati et al. [114], this can be done using $(1 - \mu_r)^{-1/N} - \phi$ curve. On this chart, which is shown in figure 4.6, the value of $\phi$ corresponding to upper limit of $(1 - \mu_r)^{-1/N} = 1$ should be identical to $\phi_m = 21.76$ which was calculated earlier. By taking $N = 1$, $\phi = 21.76$ leads to $(1 - \mu_r)^{-1/N} = 1$, which is the same as $\phi_m$. It should be noted that the proper choice for high shear plasma flows is $N = 2$. However, the value of $N =$
Figure 4.5: $1/\mu_r$ for the YSZ-water suspension. Dashed line is the trend line.

1 produces a better match to the experimental results, suggesting the experiments were performed at lower shear rates. In the calculations presented here, the choice of $N = 2$ will not result in a significant change of $\phi$ value ($\phi \approx 24$).

Figure 4.6: $(1 - \mu_r)^{-1/N}$ for the YSZ-water suspension. Dashed line is the trend line.
Figure 4.7: Effect of turbulence model on temperature profile, experimental results are from Brossa et al. [117] and numerical results from Bolot et al. [118] and Jabbari et al. [34].

Figure 4.8: Effect of mesh resolution on temperature profile, experimental results are from Brossa et al. [117].
4.3 SPS Particle Tracking

4.3.1 Torch Validation

Before continuing to particle injections, the torch model is benchmarked here against available experimental and numerical literature. For this test, a flow of Argon with 0.1 mole $H_2$ is entered the torch at 35.4 slpm. Anode diameter is 6 mm. Flow passing through anode is heated with a heat source corresponding a torch with 47% thermal efficiency, 25.6 (V), and 600 (A). The domain is discretized using 560k cells. Fig. 4.7 shows results obtained here compared to other available test cases in literature. The k-ε results are a good match to the experimental results of Brossa et al. [117] and are also close to numerical predictions of Bolot et al. [118] as well as numerical RSM results of Jabbari et al. [34]. The same test is repeated at different mesh resolutions to ensure the mesh independency. An extra fine mesh resolution (1.5M cells) and a coarser resolution (200k cells) are used for that matter. Results of these test cases
are presented in Fig. 4.8. As evident, the fine resolution of 560k cells produces close results to the extra fine resolution of 1.5M. The rest of test cases in this paper are performed at this resolution. The torch model has also been tested against experimental results of Brossa [117] for different torch operating conditions. Table 4.3 shows torch conditions for each of these test cases. Numerical results obtained here are plotted in Fig. 4.9. The torch geometry used here is slightly different from Brossas experimental setup. The outer diameter of the Brossas torch is 7.88mm against a 6mm diameter here. Brossas torch also has a diverging nozzle which is not accounted for at this time. Numerical results here, similar to other studies, manage to capture the general temperature trend of the torch and are close to the experimental data. Closer accuracies can be achieved by using better implementations of the arc, which has been left for future studies. Another test case is also performed for comparison against results reported by Meillot et al. [119]. The operating conditions are 60slpm flow rate on a torch with arc current of 500A and a voltage of 65V. The value for thermal efficiency is 50%. Results for this test case are shown in Fig. 4.10.

Table 4.3: Torch conditions

<table>
<thead>
<tr>
<th>Case #</th>
<th>I (A)</th>
<th>V (V)</th>
<th>η (%)</th>
<th>˙m(slpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>450</td>
<td>27.2</td>
<td>51.2</td>
<td>47.2</td>
</tr>
<tr>
<td>2</td>
<td>450</td>
<td>24.8</td>
<td>47.1</td>
<td>35.4</td>
</tr>
<tr>
<td>3</td>
<td>600</td>
<td>25.6</td>
<td>47.1</td>
<td>35.4</td>
</tr>
</tbody>
</table>

### 4.3.2 Effect of viscosity on breakup

The importance of using a valid viscosity model to capture suspension droplets breakup can be emphasized by examining the test case described below. Torch operating conditions of case A (Table 4.5) have been used in this test. Droplets injected are suspension of 10wt.% YSZ in water and have a constant viscosity. Ohnesorge
Figure 4.10: Velocity and temperature profile on the torch centerline against experimental and numerical results of Meillot et al. [119].

The Oh number for injected droplets as they travel from the torch towards the substrate has been plotted in Fig. 4.11. It is evident that many droplets have Oh Numbers larger than 0.1, which is the threshold from which the influence of viscosity becomes significant in the break up process [42]. With the assumption of constant viscosity, Oh numbers calculated here do not include effects of increase in viscosity as the solvent in the droplets evaporates. This neglected increase in viscosity will lead to even larger Oh numbers which will make the situation worse.

4.3.3 Effect of viscosity model

As discussed before, a number of different models may be used for predicting effective viscosity based on instantaneous volume fraction of solids in suspension droplets. Since the experimental result for viscosity of 8 mol % YSZ suspension in water at different concentrations is available (Fig. 4.4), it has been used as a benchmark for the models described here. In one case, the experimental values for viscosity
Chapter 4. Suspension Plasma Spraying (SPS)

Figure 4.11: Ohnesorge Number versus flight time for YSZ-water suspension particle injected into the domain.

of this suspension have been used to update suspension viscosity at various solid concentrations. In this manner, at each time step in simulation, the volume fraction of solid content in each injected flying droplet is recalculated. This value is then used to update the viscosity for the suspension mixture of that particular droplet. This process is repeated for all injected droplets and during all iterations. Keeping all conditions identical, the same test case is repeated here, but instead, the viscosity of the suspension mixture is updated using equations 4.27, 4.28, 4.29, 4.31, and 4.33. Equation 4.32 is not examined as suspension powder size distribution is neglected in the current model. Viscosity predictions from each equation has been plotted in Fig. 4.12 for different volume fraction values. Values close to $\phi_m$ have not been plotted, as predictions start growing to different large values, all being estimates for infinitely large viscosity. This figure suggests predictions by equation 4.33 are closer to experimental values of Fig. 4.4 compared to the rest of equations.

Simulation results for these test cases have been summarized in Fig. 4.13. Torch op-
erating conditions here are that of case C in Table 4.5. Particles traveling away from torch are here collected on a substrate located 8cm downstream of the nozzle exit. This figure shows the effect of viscosity model on the probability density distribution of particle diameter and Reynolds number upon impact on the substrate. Updating particle viscosities using equation 4.33 produces closer results to experimental viscosity data of Fig. 4.4. Equations 4.27, 4.28, 4.29, and 4.31 result in larger errors. Equation 4.28 is the least accurate.

4.3.4 Effects of Injection Parameters

In the following section, effects of injection parameters on the particle trajectories are investigated. Droplets are injected with the assumption of being already atomized. For the moment, the size distribution of injected droplets is neglected and it is assumed that all droplets have the same diameter. Injection is controlled by four parameters: (i) Velocity magnitude of droplets at injection time ($v$), (ii) Injection angle ($\theta$), (iii) Height ($H$), which is the injectors distance from the torch exit, and
Figure 4.13: Particle standardized diameter and Reynolds number density distribution for different viscosity models.
(iv) Radial distance ($R$), which is the injectors distance from the centreline of the torch. A schematic drawing of these parameters is shown in Fig. 4.14.

![Figure 4.14: Schematic drawing of the parameters defining injection conditions.](image)

After injection, droplets are tracked and undergo breakups, evaporation, or they may coalesce with one another. The liquid phase evaporates during flight and then solid phase starts melting. To save computational time, radiation heat losses are not considered for the test cases in current section. Molten particles are carried towards the substrate. In place of the substrate, 8cm downstream of the torch, a wall is placed to capture the particles impacting the substrate. The torch used for the calculations here has a nozzle diameter of 6mm, thermal efficiency of 47%, and is operated at 25.6 ($V$) and 600 ($A$). For Ar-H2 gas mixture, according to [94], the accommodation coefficient ($a$) and in equation 4.12 are assumed to be 0.8 and 1.411, respectively.

For each test case in Table 4.4, particles are captured on a substrate that is located
8 cm from the nozzle exit. The spraying mass flow rate for all cases is kept the same. This means the time step for these test cases are different. The number of particles impacted the substrate during 1000 iterations is shown in Fig. 4.15. Analysis of the particle trajectories is of interest here. We may examine how the injection parameters have led to particle penetration through the plasma jet. For many of the test cases, it is evident that injection parameters have caused the particles to miss the large substrate. This clearly shows the position of the torch and the mass flow rate of the suspension play important role in efficiency. Test cases 28-54, possess an overall larger particle count on the substrate compared to the rest of cases. This suggest that lowering the injection site closer to the torch centerline ($R = 0$) will help improving particle counts on the substrate. This however may not be feasible for some torches and injectors. The results also show that increasing the injection velocity of particles, can work in two ways. For instance, keeping the rest of variables the same, increasing the velocity from 10 to 20 m/s for test cases 13 and 14 has led to fewer particles reaching the substrate. The increase of velocity to 40 in case 15, has caused a complete penetration of the suspension particles and hence none have ended on the desired position. The same pattern can also be observed for cases 10-12, 19-21, 22-24, 46-18, and 49-51. For cases 4-9, 16-18, and 25-27 the injection is aimed too deep in the torch and hits inner and outer parts of the torch geometry and is scattered before successfully entering the main flow path. The largest number of particle count is related to case 37. This was predictable as in this case, the injection site is located on the centerline. Also the velocity of particles at injecting time is small compared to other cases, and hence gives the particles less chance for escaping the core flow. Test case 28 has also similar specification to case 37. The only difference is the injection site is moved from 1 cm to 5 mm from the nozzle exit. The moving of the injection site closer to the exit here has had a negative impact on the particle paths. This
may indicate that the torch has recovered its symmetry better when the injection point was located at 1cm compared to 5mm. Moving the injection site downstream at 1.5cm, as in case 46, has also decreased the particle counts. This indicates the 1cm standoff is possibly closer to the ideal position of the injection site on the torch centreline.

Figure 4.15: Particle counts on substrate for each test case.

### 4.3.5 Effect of Torch Operating Conditions

Plasma torch operating conditions play a key role in the effectiveness of the SPS process. Table 4.5 shows the range of operating conditions for the torch considered in this work, which include the Ar-H2 mass flow rate and arc voltage variations. For case A, B, and C, only the gas flow rate is varied. Torch power is also varied using the voltage value. For cases A, D, and E, the change in voltage generates power variations of 7,235W, 14,470W, and 21,704W, respectively. The mass Flow rate for these three cases is kept identical (35.4slpm). In all test cases, the right boundary of
## Table 4.4: Test case conditions

<table>
<thead>
<tr>
<th>Case #</th>
<th>R (m)</th>
<th>H (m)</th>
<th>θ (deg)</th>
<th>v (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.025</td>
<td>0.005</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>2</td>
<td>0.025</td>
<td>0.005</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>3</td>
<td>0.025</td>
<td>0.005</td>
<td>10</td>
<td>40</td>
</tr>
<tr>
<td>4</td>
<td>0.025</td>
<td>0.005</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>5</td>
<td>0.025</td>
<td>0.005</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>6</td>
<td>0.025</td>
<td>0.005</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>7</td>
<td>0.025</td>
<td>0.005</td>
<td>40</td>
<td>10</td>
</tr>
<tr>
<td>8</td>
<td>0.025</td>
<td>0.005</td>
<td>40</td>
<td>20</td>
</tr>
<tr>
<td>9</td>
<td>0.025</td>
<td>0.005</td>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>10</td>
<td>0.025</td>
<td>0.01</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>11</td>
<td>0.025</td>
<td>0.01</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>12</td>
<td>0.025</td>
<td>0.01</td>
<td>10</td>
<td>40</td>
</tr>
<tr>
<td>13</td>
<td>0.025</td>
<td>0.01</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>14</td>
<td>0.025</td>
<td>0.01</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>15</td>
<td>0.025</td>
<td>0.01</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>16</td>
<td>0.025</td>
<td>0.01</td>
<td>40</td>
<td>10</td>
</tr>
<tr>
<td>17</td>
<td>0.025</td>
<td>0.01</td>
<td>40</td>
<td>20</td>
</tr>
<tr>
<td>18</td>
<td>0.025</td>
<td>0.01</td>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>19</td>
<td>0.025</td>
<td>0.015</td>
<td>10</td>
<td>10</td>
</tr>
<tr>
<td>20</td>
<td>0.025</td>
<td>0.015</td>
<td>10</td>
<td>20</td>
</tr>
<tr>
<td>21</td>
<td>0.025</td>
<td>0.015</td>
<td>10</td>
<td>40</td>
</tr>
<tr>
<td>22</td>
<td>0.025</td>
<td>0.015</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>23</td>
<td>0.025</td>
<td>0.015</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>24</td>
<td>0.025</td>
<td>0.015</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>25</td>
<td>0.025</td>
<td>0.015</td>
<td>40</td>
<td>10</td>
</tr>
<tr>
<td>26</td>
<td>0.025</td>
<td>0.015</td>
<td>40</td>
<td>20</td>
</tr>
<tr>
<td>27</td>
<td>0.025</td>
<td>0.015</td>
<td>40</td>
<td>40</td>
</tr>
<tr>
<td>28</td>
<td>0</td>
<td>0.005</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>29</td>
<td>0</td>
<td>0.005</td>
<td>0</td>
<td>20</td>
</tr>
<tr>
<td>30</td>
<td>0</td>
<td>0.005</td>
<td>0</td>
<td>40</td>
</tr>
<tr>
<td>31</td>
<td>0</td>
<td>0.005</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>32</td>
<td>0</td>
<td>0.005</td>
<td>15</td>
<td>20</td>
</tr>
<tr>
<td>33</td>
<td>0</td>
<td>0.005</td>
<td>15</td>
<td>40</td>
</tr>
<tr>
<td>34</td>
<td>0</td>
<td>0.005</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>35</td>
<td>0</td>
<td>0.005</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>36</td>
<td>0</td>
<td>0.005</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>37</td>
<td>0</td>
<td>0.01</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>38</td>
<td>0</td>
<td>0.01</td>
<td>0</td>
<td>20</td>
</tr>
<tr>
<td>39</td>
<td>0</td>
<td>0.01</td>
<td>0</td>
<td>40</td>
</tr>
<tr>
<td>40</td>
<td>0</td>
<td>0.01</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>41</td>
<td>0</td>
<td>0.01</td>
<td>15</td>
<td>20</td>
</tr>
<tr>
<td>42</td>
<td>0</td>
<td>0.01</td>
<td>15</td>
<td>40</td>
</tr>
<tr>
<td>43</td>
<td>0</td>
<td>0.01</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>44</td>
<td>0</td>
<td>0.01</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>45</td>
<td>0</td>
<td>0.01</td>
<td>20</td>
<td>40</td>
</tr>
<tr>
<td>46</td>
<td>0</td>
<td>0.015</td>
<td>0</td>
<td>10</td>
</tr>
<tr>
<td>47</td>
<td>0</td>
<td>0.015</td>
<td>0</td>
<td>20</td>
</tr>
<tr>
<td>48</td>
<td>0</td>
<td>0.015</td>
<td>0</td>
<td>40</td>
</tr>
<tr>
<td>49</td>
<td>0</td>
<td>0.015</td>
<td>15</td>
<td>10</td>
</tr>
<tr>
<td>50</td>
<td>0</td>
<td>0.015</td>
<td>15</td>
<td>20</td>
</tr>
<tr>
<td>51</td>
<td>0</td>
<td>0.015</td>
<td>15</td>
<td>40</td>
</tr>
<tr>
<td>52</td>
<td>0</td>
<td>0.015</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>53</td>
<td>0</td>
<td>0.015</td>
<td>20</td>
<td>20</td>
</tr>
<tr>
<td>54</td>
<td>0</td>
<td>0.015</td>
<td>20</td>
<td>40</td>
</tr>
</tbody>
</table>
the domain at 8cm is a large substrate.

Table 4.5: Effect of torch parameters

<table>
<thead>
<tr>
<th>Case #</th>
<th>I (A)</th>
<th>V (V)</th>
<th>η (%)</th>
<th>P (W)</th>
<th>m (slpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>600</td>
<td>25.6</td>
<td>47.1</td>
<td>7,235</td>
<td>35.4</td>
</tr>
<tr>
<td>B</td>
<td>600</td>
<td>25.6</td>
<td>47.1</td>
<td>7,235</td>
<td>70.0</td>
</tr>
<tr>
<td>C</td>
<td>600</td>
<td>25.6</td>
<td>47.1</td>
<td>7,235</td>
<td>140.0</td>
</tr>
<tr>
<td>D</td>
<td>600</td>
<td>51.2</td>
<td>47.1</td>
<td>14,470</td>
<td>35.4</td>
</tr>
<tr>
<td>E</td>
<td>600</td>
<td>76.8</td>
<td>47.1</td>
<td>21,704</td>
<td>35.4</td>
</tr>
</tbody>
</table>

In all these cases, the injected suspension is 10 wt% YSZ-water. Droplets are injected with a uniform initial diameter of 150μm. The injector properties for these test cases are \( R=0.005\text{m} \), \( H=0.005\text{m} \), \( \theta = 10^\circ \), and \( v=10\text{m/s} \). Injection flow rate is kept constant at 1.92E-4kg/s. The suspension viscosity is corrected utilizing Fig. 4.4. Evaporation of water, breakup and coalescence of liquid suspension and molten drops, and radiation heat losses are all taken into account. Two particular aspects affecting the droplets/particles are of interest. First is the conditions during its flight, e.g., shear forces exerted on the droplets at each particular point of their flight path. The second area of interest is particle conditions upon impact on the substrate. Conditions such as particle temperature, diameter, and velocity play an important role in defining the final finish of the coating. In the following sections, for test cases A-E, initially droplet conditions during flight are examined. Afterwards, particle properties upon impact on the substrate are investigated.

4.3.6 In-flight conditions

The injection of liquid suspension alters the flow pattern of the plasma gas. The momentum carried by the liquid jet traveling towards the torch divides the flow into two steams for a short distance. Temperature contours on axial cross sections of
the torch from two perpendicular angles have been shown in Fig. 4.16 for case A. Images on the left show the torch with injection and substrate, while images on the right show the torch without injection and substrate. The liquid jet alters the torch flow pattern by slightly shifting the position of the maximum velocity at the nozzle exit. Also the liquid injected is typically at room temperature. This has a cooling effect on the plasma flow. More importantly, evaporation of the liquid content in the suspension is the major cause of cooling of the plasma.

As another example, temperature profile of the plasma flow for case E has been plotted in Fig. 4.17 on the centerline of the torch. This chart clearly shows the cooling effects of suspension on plasma. It can be seen that the flow near the injection penetration site has a sudden drop in temperature, which starts recovering after 5mm downstream where the flow symmetry is being restored. Fig. 4.18 shows the temperature field for case A at three cross sections, with distances of 5, 7, and 9mm from the nozzle exit. Images on the left are with injection, while images on the right are without injection. The injector here is aimed close to the center point of the nozzle exit. It is evident that due to existence of the liquid stream, the temperature profile is no longer symmetric.
at each cross section. Comparison of the temperature profiles shows the important cooling effect. This cooling is not significant before 5mm. However, as the liquid phase evaporates, the cooling becomes more pronounced, as can be seen in the cross section at 9mm. Similar to findings of Fazilleau et al. [31], the torch here recovers its symmetry flow pattern after 1-2cm downstream of the nozzle exit. The maximum temperature of the flow however is lower after the symmetry is restored compared to when there is no injection. Fig. 4.19 shows a snapshot of particle positions during their flight for cases A-E.

To study the effect of plasma-particle interactions more closely, other properties of particles for case A have been plotted in Fig. 4.20. These properties are the diameter of each particle (D), mass fraction of water inside each particle that has not been evaporated yet, particle Reynolds number defined as $Re = \rho_g v_{rel} d_p / \mu_g$, particle Weber number defined as $We = \rho_g v_{rel}^2 d_p / \sigma_p$, particle Ohnesorge number defined as $Oh = \mu_p / \sqrt{\rho_p \sigma_p d_p}$, and shear/strain rate at the cell where the particle is located. The
Figure 4.18: Temperature profile for case A, plotted at cross sections perpendicular to the flow direction at distances of 3, 5, and 7mm from the nozzle exit (from top to bottom).
Figure 4.19: Snapshot of particle positions during flight for Cases A-E. Images on the right show particles during their flight from substrates point of view. Particles are colored based on their diameter (m).
Chapter 4. Suspension Plasma Spraying (SPS)

x-axis here indicates the distance each particle has travelled from the nozzle exit.

Figure 4.20: Diameter, velocity, Reynolds number, Weber number, Ohnesorge number, and shear rates for particles during flight. Shown here for case A. The torch nozzle exit is located at X=0.02m.

The Reynold and Weber diagrams of the droplets exhibits a huge jump near the nozzle exit and converges fast to smaller values as they move away from the nozzle exit. The ratio of these two has been plotted as the Ohnesorge number. The values of the Oh become large, especially after the liquid content is evaporated. This increase in Oh should be handled with caution as the apparent increase in viscosity, imposed here to numerically mimic the presence of solid YSZ particles, might be contributing to it. Hence, the values of these non-dimensional numbers must be studied next to particle temperatures. For case A, particles have temperature readings larger than
melting point from approximately 15 to 33\textit{mm}. Many particles are also at their melting temperature and might be in the mushy region from 18 to 51\textit{mm}. Over this range, many particles have Reynolds numbers below 3. Small groups also have values close to 30, 50, and 150. The Weber numbers are all below 7 at this range, with many having values below 0.01. These values result in two Ohesorge numbers close to 0.3 and 3. Shear rate trends can also be seen in Fig. 4.20. These trends are close for all test cases. The maximum is similar for test cases A and C. Cases B and E have slightly higher rates, while the largest is seen for case D. In these figures, small number of droplets can be observed having very low shear rates. These are the droplets that have either penetrated through the plasma jet flow at the injection point, or have escaped the flow and are travelling far from the centerline. If the injection parameters are not chosen correctly, the liquid suspension can penetrate past the plasma jet and many of the droplets will end up escaping nearly untouched by not going through evaporation and breakups. The liquid mass fractions inside droplets during flight is shown in Fig. 4.21 for cases A to E. The torch nozzle exit is located at \( X = 0.02\text{m} \) here. Fazilleau et al. [31] have reported that at 10 – 15\textit{mm} downstream of the nozzle exit, all the liquid content in the suspension will be already evaporated. For test cases here, the liquid content in most of the injected droplets is evaporated nearly at that distance. For case C however, the evaporation of solvent takes longer. Compared to cases A and B, case C has the same amount of power, but also a higher mass flow rate. Hence the particles injected into this flow will have less time to absorb enough energy to finish their phase change cycle. Best performance here is seen for case E, where all particles lose their liquid solvent and at the fastest rate.

Temperature profile for the cases A and E have been compared in Fig. 4.22. This snapshot of particles during flight shows that whether the particles have melted or
Figure 4.21: Water mass fraction inside particles during flight for cases A-E.
not, still have their maximum temperature at 15 to 20\( \text{mm} \) from nozzle exit. After this point, the temperature starts to drop. This is mainly due to the fact that the plasma flow, as seen in Fig. 4.17, has started to cool down close to this point. The rise in particle temperatures also lowers the rate of heat transfer. Particles now being at a higher temperatures will dissipate heat more rapidly through radiation. As will be seen later, from the test cases here, more particles in cases D and E have temperature values higher than melting point of YSZ. This suggests the increase of power from 7kW to 14kW has a direct impact on better evaporation of liquid phase and melting of the solid YSZ particles. Temperature profiles of particles are nearly the same for cases D and E which indicates that the rise of power from 14kW to 21kW does not significantly increase the inflight maximum temperatures.

4.3.7 Substrate Collections

Another important location for obtaining particle variables is on the substrate. Fully molten particles travelling at different speeds will generate different surface finishes. Moreover, under certain conditions, there is the possibility of having unmolten, semi molten, or even wet mixtures impacting the substrate. For the test cases here, a large substrate is placed 8cm downstream of nozzle exit. This surface will generate a stagnation flow pattern which helps in understanding of the particle interactions with the substrate. In previous studies, particles are typically collected in a region near the substrate. Here however, particles are only collected upon impact on the substrate. The rest of particles that come close to the wall but manage to escape are not included in here. It is also assumed that particles hitting the surface will adhere to it and do not rebound. The temperature distribution for the particles captured on the substrate for cases A-E are plotted in Fig. 4.23. As the inlet flow rate of
Figure 4.22: Temperature of particles during flight for cases A and E.
$Ar-H_2$ changes from $35 \text{slpm}$ for case A to $70 \text{slpm}$ for case B and $140 \text{slpm}$ for case C, the peak of the particle temperatures is shifted to lower temperature values. This shows that by keeping the torch power constant, the higher flow of gas has generated cooling effects which eventually has resulted in lower particle temperature. Particles travelling at higher speeds have less time for proper heat transfer. For these cases, the liquid content in some of the particles has not been fully evaporated by the time they reach the substrate. This is shown in the small peak below $500K$, close to the evaporation temperature of water. As expected, the number of wet particles for case C is larger than cases A and B.

![Particle temperature distributions for test cases A-E upon impact on the substrate.](image-url)
Particles impacting the substrate for cases D and E have higher temperatures. For case D, most of the particles are semi-molten, explained by the sharp peak near the melting point of YSZ. This means that energy of the torch at 14kW was only enough to fully evaporate liquid phase and increase particle temperatures to the melting point. However, it was insufficient to help particles go through their melting process. Fig. 4.24 shows particle diameter distributions for all test cases as they impact the substrate. These values are the final diameters of the particles after all the evaporations and breakups have taken place. Results show the increase of mass flow from case A to C shifts the peak to lower diameter sizes. The change from $35\text{slpm}$ to $70\text{slpm}$ seems to have a more significant effect compared to the change from $70\text{slpm}$ to $140\text{slpm}$. The increase of the mass flow to $140\text{slpm}$ has only made the diameter range slightly narrower. Near the values of $70 - 140\text{slpm}$, most particles have diameters close to $3\mu m$. The diameter for particles here are slightly larger than previously reported experimental values. This can be explained since the scattering and explosion of solid YSZ particles are not included in the assumptions. In reality, the solid particles inside the suspension do not form one sphere and can decrease the final size distribution of droplets.

Increasing the power from test case A to D and E also affects the final particle diameters. Comparing the distribution of diameters between these three cases shows that the particles for all three cases nearly cover the same range of diameters. By increasing the power, however, the distributions have become less spread. For case A, for instance many particles reaching the substrate are still wet, carrying unevaporated water. These particles have gone through several stages of breakup due to shear force exerted by plasma flow. However, these suspensions have not managed to absorb sufficient heat to lose their moisture. Fig. 4.25 shows the particles from test case B that have been captured on the substrate over a certain time period. This figure
Figure 4.24: Particle diameter distributions for test cases A-E upon impact on the substrate.
shows that the torch footprint has slightly shifted to the left. This is explained by the fact that the suspension was injected from right to left.

Figure 4.25: Particles captured on the substrate 8cm downstream of torch for test case B collected over 3ms.

The cumulative velocity distributions for particles upon impact on the substrate have been plotted in Fig. 4.26. The velocity of particles has been divided into two components: one is the normal velocity which is the velocity component perpendicular to
the substrate, and the other is the tangential velocity which is the velocity component parallel to the substrate. In an ideal situation, all momentum energy in the particles should be invested in a normal impact, making the tangential velocities zero. This is however impossible, as the stagnation flow pattern near the substrate will force particles to lose normal momentum. Case A here has lower tangential velocities. Increasing the power and mass flow rate have both led to higher velocities. The tangential velocities for cases B, C, and E have a similar pattern. The normal velocities here for all cases cover the same velocity ranges. Particles travelling close to the torch center line maintain higher velocities. As a typical substrate is smaller than the plane of study here, it can be predicted that most of the particles impacting the substrate will have the higher velocity ranges. However, for when the torch is not stationary, or the substrate of interest is large, the particles with smaller velocities will also impact the substrate. Different velocities upon impact will result in different splat sizes which will affect the coating microstructure. The normal velocity values follow nearly the same trend for all test cases. The line for test case B is higher than other cases at low velocities. Examining particle positions when impacting the substrate for case B reveals that particles at velocities higher than 25 m/s have ended in a circle in the middle of the substrate. Particles with lower velocities however have covered a larger radius. Diameter measurements for both fast and slow particles are nearly the same. All particles with lower velocities also have lower temperature. This suggests that for this test case, the particles have been travelling close to the main plasma flow. These particles were unable to escape the main flow pattern, yet were not close enough to the centerline in order to receive enough heat. For the rest of test cases, it can be observed that the rise in power from 7kW to 21kW has clearly given particles higher impact velocities.
Chapter 4. Suspension Plasma Spraying (SPS)

Figure 4.26: Particle normal and tangential velocity distributions for test cases A-E upon impact on the substrate.
Chapter 5

SPS Droplet Impact & Solidification

5.1 Problem Specification

The information gathered before can now be used to capture impact of generated YSZ particles on the substrate using the SPH formulation. From test cases reviewed before, sample particles have been chosen for closely monitoring their impacts on the substrate. Analyzing impact points of droplets from previous section reveals that only a handful will have close impact points on the substrate. This comes from high efficiency of the SPS process in coating the surface, as the mist of particles generated from atomization of liquid suspension uniformly covers a large area, making it nearly impossible to get multiple impacts on a particular place on the substrate in a short time interval. Here, a site on the substrate has been chosen for further study, where SPS data shows close impact of 5 particles (from case E, table 4.5). The impact points of these 5 particles are shown in figure 5.1. Although not all of these 5 particles have direct impacts, they will eventually overlap when their splats spread over the

122
substrate. Properties of these particles at the time of impact are provided in table 5.1. As evident, droplets in this neighborhood cover a wide range of possible diameters that are found during SPS coating.

![Position and diameter of five YSZ drops impacting the substrate obtained from case E (table 4.5).](image)

Figure 5.1: Position and diameter of five YSZ drops impacting the substrate obtained from case E (table 4.5).

<table>
<thead>
<tr>
<th>#</th>
<th>x (µm)</th>
<th>y (µm)</th>
<th>u (m/s)</th>
<th>v (m/s)</th>
<th>w (m/s)</th>
<th>T (K)</th>
<th>D (µm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>-6.6</td>
<td>3.0</td>
<td>-4.1</td>
<td>1.5</td>
<td>-207.6</td>
<td>3679.8</td>
<td>9.1</td>
</tr>
<tr>
<td>2</td>
<td>1.4</td>
<td>-2.9</td>
<td>-0.2</td>
<td>5.9</td>
<td>-226.1</td>
<td>3833.5</td>
<td>8.7</td>
</tr>
<tr>
<td>3</td>
<td>10.8</td>
<td>-9.3</td>
<td>27.7</td>
<td>5.1</td>
<td>-88.5</td>
<td>3523.5</td>
<td>3.0</td>
</tr>
<tr>
<td>4</td>
<td>4.7</td>
<td>5.7</td>
<td>15.3</td>
<td>16.1</td>
<td>-99.3</td>
<td>3543.3</td>
<td>2.9</td>
</tr>
<tr>
<td>5</td>
<td>-10.3</td>
<td>3.6</td>
<td>-8.1</td>
<td>35.3</td>
<td>-227.8</td>
<td>3638.2</td>
<td>4.2</td>
</tr>
</tbody>
</table>

Table 5.1: YSZ particles properties at impact
In figure 5.1, the substrate is located in the $x - y$ plane. The $z$ axis is perpendicular to the substrate and points towards the torch nozzle. Particles traveling to the substrate will have a negative velocity in the $z$ direction. Velocity components parallel to the substrate, though small in magnitude, have also been included in this simulation. Sample simulation cases show that the impact, spread and solidification of each particle happens orders of magnitude faster than the impact of the next particle. Hence, it is reasonable to assume that each particle is completely solidified before the next particle impacts close to it. For the simulations here, the material for the droplets and substrate is YSZ. Though at the start of the coating process, the substrate material will play an important role as typical metallic substrates will have higher thermal conductivities compared to YSZ, with more YSZ layers coating the surface over time, the new droplets will mostly impact a substrate covered by YSZ. The droplets are in molten liquid states corresponding to their values in table 5.1. The contact angle between drops and the surface is taken to be $90^\circ$ [83], though as it will be discussed later, the capillary effects are negligible since $We \gg \sqrt{Re}$ here [120]. The solidification of molten drops is resolved using equation 2.49. The mushy zone effects are also included using equation 2.77. Gravity force is added with a value of $9.81 m/s^2$ exerted on molten particles in the opposite direction of the $z$ axis, although its effect is also not significant. As the impact velocities are high, it is important to apply a no-slip boundary condition on the substrate. Here, the efficient no-slip model of Holmes et al. [48] has been utilized which performs well on both flat and more complex wall geometries.
5.2 Multiple Splat Overlaps

Results are presented in figures 5.2-5.8. Figures have been colored by $z$ magnitude at each point, identifying how the roughness of the substrate changes due to the increase in local height. Figures 5.2, 5.3, 5.5, and 5.6 show substrate after impact of droplets #1, #2, #3, and #4, respectively. The first two droplets have larger diameters near 9$\mu$m with impact velocities higher than 200$m/s$. Hence, they have large spread factors and the splats cover a large area. The next two drops have smaller diameters near 3$\mu$m and lower impact velocities below 100$m/s$. The last drop also has a small diameter, but impacts at the highest speed. The final coated substrate after five impacts is plotted in figures 5.7 and 5.8. The maximum height from substrate’s surface for each solidified droplet is 0.91, 1.76, 1.53, 2.40, and 2.14 $\mu$m, for droplets #1 to #5, respectively. Hence, the highest peak in the finished coating corresponds to a height of 2.40$\mu$m, which is achieved from overlapping splats of droplets #1, #2, and #4.

5.3 Spread Factor

As impact points of YSZ particles generated by suspension plasma spraying are well-spread on the substrate, the larger size droplets make the main contribution in creating roughness, by having splats that overlap one another. Smaller size particles mostly create isolated bumps, as the possibility of having direct binary impacts in short time periods is low, and because the spread factor for these smaller droplets is not relatively large. Table 5.2 shows some statistical information on YSZ particles in case E. For different particle diameter ranges, the number of YSZ droplets, the average impact velocities, and the average distance between the impact sites and
Figure 5.2: Solidification of droplet #1 on the substrate (table 5.1) colored by the height from the substrate.
Figure 5.3: Solidification of droplet #2 on the substrate (table 5.1) colored by the height from the substrate.
Figure 5.4: Solidification of droplet #2 on the substrate (table 5.1) colored by the height from the substrate.
Figure 5.5: Solidification of droplet #3 on the substrate (table 5.1) colored by the height from the substrate.
Figure 5.6: Solidification of droplet #4 on the substrate (table 5.1) colored by the height from the substrate.
Figure 5.7: Solidification of five drops on the substrate (table 5.1) colored by the height from the substrate.
Figure 5.8: Solidification of five drops on the substrate (table 5.1) colored by the height from the substrate.
the center of all impacts ($\bar{r}$) have been reported in this table. Here, $r$ is defined as $\sqrt{(x - x_{\text{mean}})^2 + (y - y_{\text{mean}})^2}$. $x_{\text{mean}}$ and $y_{\text{mean}}$ mark the mean center for all the impacts. While the torch is located and is pointing to (0, 0), since the injection is done with an angle, the particles will not impact near the (0, 0) point and will be shifted to the direction imposed by injection angle.

It is evident that small size ranges below 3 $\mu m$ exhibit smaller impact velocities (smaller $\bar{w}$). These droplets are also falling further from the center of the substrate (larger $\bar{r}$). Larger YSZ droplets, on the other hand, have filled an area with diameter of 10 mm, potentially influenced by the torch nozzle diameter which is 7 mm. Combining findings from impact studies with the statistics here, it can be estimated that during this spray time, the substrate would possess a rougher finish inside the 10 mm circle, which results from large particles imparting and generating overlapping of splats. The overlap of the two large particles in the test case conducted here (particles #1 and #2 in Table 5.1) generated a height of 1.76 $\mu m$. The SPH simulation of drop #1 impact shows a spread factor ($D/D_0$) of 3.

Table 5.2: Average impact velocity ($\bar{w}$) and distance from substrate center ($\bar{r}$) for different diameter ranges in case E (Table 4.5).

<table>
<thead>
<tr>
<th>$D$ ($\mu m$)</th>
<th>count (%)</th>
<th>$\bar{w}$ (m/s)</th>
<th>$\bar{r}$ (mm)</th>
<th>$\text{max}(r)$ (mm)</th>
<th>$\text{min}(r)$ (mm)</th>
<th>$\bar{T}$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-2</td>
<td>1</td>
<td>43</td>
<td>10.46</td>
<td>35.66</td>
<td>1.38</td>
<td>3142</td>
</tr>
<tr>
<td>2-3</td>
<td>7</td>
<td>80</td>
<td>7.73</td>
<td>37.56</td>
<td>0.10</td>
<td>3330</td>
</tr>
<tr>
<td>3-4</td>
<td>13</td>
<td>131</td>
<td>5.84</td>
<td>31.27</td>
<td>0.25</td>
<td>3468</td>
</tr>
<tr>
<td>4-5</td>
<td>16</td>
<td>169</td>
<td>5.75</td>
<td>39.41</td>
<td>0.09</td>
<td>3522</td>
</tr>
<tr>
<td>5-6</td>
<td>17</td>
<td>197</td>
<td>5.37</td>
<td>21.59</td>
<td>0.11</td>
<td>3591</td>
</tr>
<tr>
<td>6-7</td>
<td>16</td>
<td>211</td>
<td>5.33</td>
<td>30.39</td>
<td>0.13</td>
<td>3656</td>
</tr>
<tr>
<td>7-8</td>
<td>12</td>
<td>213</td>
<td>5.31</td>
<td>16.34</td>
<td>0.10</td>
<td>3690</td>
</tr>
<tr>
<td>8-9</td>
<td>8</td>
<td>199</td>
<td>5.64</td>
<td>15.14</td>
<td>0.31</td>
<td>3673</td>
</tr>
<tr>
<td>9-10</td>
<td>7</td>
<td>194</td>
<td>5.33</td>
<td>15.66</td>
<td>0.29</td>
<td>3717</td>
</tr>
<tr>
<td>10-11</td>
<td>3</td>
<td>179</td>
<td>5.49</td>
<td>12.55</td>
<td>0.39</td>
<td>3685</td>
</tr>
<tr>
<td>11-12</td>
<td>1</td>
<td>204</td>
<td>5.53</td>
<td>8.54</td>
<td>2.30</td>
<td>3905</td>
</tr>
</tbody>
</table>
Scatterness of particles on the substrate has been shown in figures 5.9 and 5.10 for different particle diameters. Results show for particle sizes between 1 to 3µm the particles cover a larger area on the substrate with a radius near 3.5 to 4cm. Particles get closer to the center as the sizes grow higher.

Figure 5.9: Distribution of impact position classified based on particle diameter.

To get a better prediction on the final conditions of the substrate, the SPH solver has been used here to calculate spread factor at three different velocities of 50, 100, and 200m/s, which covers the velocity range of YSZ droplets impacting the substrate obtained from SPS simulation. Results of this test case are plotted in figure 5.11 as a function of the non-dimensional time ($t^* = tV/D_0$). These configurations leads to non-dimensional values of 96, 192, 383 for $Re = \frac{\rho V_0 D_0}{\mu}$, and 323, 1293, 5172 for
Figure 5.10: Distribution of impact position classified based on particle diameter.
\[ We = \frac{\rho V^2 D_0}{\sigma}. \] Since Weber numbers here are much larger compared to Reynolds values, i.e. \( We \gg \sqrt{Re} \), the capillary effects can be neglected [120]. Pasandideh-Fard et al. [120] have predicted that for these conditions and with \( We \gg 12 \), spread factor can be approximated by \( 0.5Re^{0.25} \). Results obtained here also suggest a similar correlation for spread factor in the form of \( (R^2 = 0.9960) \)

\[ \xi_{\text{max}} = \frac{D_{\text{max}}}{D_0} = 0.59 \ Re^{0.28} \]  

(5.1)

Figure 5.11: Spread factor \( (D/D_0) \) for a YSZ droplet impacting the substrate at 50, 100, and 200 m/s \( (t^* = tV/D_0) \).

### 5.4 Solidification Time and Heat Flux

A test case is conducted here to examine the solidification time and amount of heat transferred from an individual molten YSZ droplet to the substrate. A droplet with diameter of 3\( \mu \text{m} \) impacts the substrate at 3000K and velocity of 150m/s. The substrate is at 600K. The substrate is chosen from two different materials. The first is YSZ, identical to the material inside the droplets. For the second, thermal conductivity of
Table 5.3: An estimate of the number of splat overlaps after impact on the substrate.

<table>
<thead>
<tr>
<th># overlaps</th>
<th>count (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>59.61</td>
</tr>
<tr>
<td>1</td>
<td>22.19</td>
</tr>
<tr>
<td>2</td>
<td>9.99</td>
</tr>
<tr>
<td>3</td>
<td>4.61</td>
</tr>
<tr>
<td>4</td>
<td>2.04</td>
</tr>
<tr>
<td>5</td>
<td>0.95</td>
</tr>
<tr>
<td>6</td>
<td>0.35</td>
</tr>
<tr>
<td>7</td>
<td>0.17</td>
</tr>
<tr>
<td>8</td>
<td>0.06</td>
</tr>
<tr>
<td>9</td>
<td>0.03</td>
</tr>
<tr>
<td>10</td>
<td>0.01</td>
</tr>
<tr>
<td>11</td>
<td>0.00</td>
</tr>
</tbody>
</table>

the substrate is chosen to be $66\text{W}/(m \cdot K)$ (similar to Tin), mimicking initial layer of coating where droplets hit a metallic substrate. Figure 5.12 shows this droplet at various stages of impact and solidification at non-dimensional times of $t^* = 0.5, 10, 500, 1000, 1400, \text{and } 1500\text{ on the YSZ substrate. Solidification starts from lower parts of the splat, and grows faster near the splat edges and in the middle, potentially due to lower thicknesses and longer contact time with the substrate, respectively. The splat fully solidifies here by nearly } t^* = 1500.$

Figure 5.13 shows the amount of heat flux at the impact point of the droplet on the substrate with higher thermal conductivity as a function of non-dimensional time. Results show that moments after impact, the heat transfer to the substrate grows suddenly to a very large value. The reason behind this sudden jump is that at the moment of impact, the substrate and droplet are at their highest temperature difference. Moreover, the impact area between these two is at its smallest near the impact time. As the droplet hits the surface and expands, the area underneath grows and distribute the heat transfer underneath the droplet. As the droplet spreads and
Figure 5.12: Impact and solidification time for a droplet at temperature of 3000K with radius of 3µm and velocity of 150m/s.
loses heat, the temperature difference between the substrate and droplet becomes smaller causing a decrease in heat flux values. The high thermal conductivity of the substrate here contributes to a fast formation of a solidified layer next to the boundary. The cooling process of the material in the bulk of splat is still limited by the low thermal conductivity of the YSZ.

Another case of importance comes from examination of YSZ droplets obtained from SPS data. Results indicate that many of the binary impacts of splats occurring on the substrate involve $4 - 6\,\mu m$ droplets. This is due to the fact that these droplets are large enough to create big splats that can effectively contribute to overlaps and surface coverage. In this manner, a test is performed here for impact of a candidate droplet from SPS data with a diameter of $4.59\,\mu m$ impacting the substrate at $175\,m/s$ with a temperature of $3517\,K$. The solidified splat for this case is shown in figure 5.14. The impact and spread of this drop on the substrate is finished by nearly $t^* = 110$. It then takes up to $t^* = 1500$ for the splat to fully solidify.

From the binary impacts involving the droplet shown in figure 5.14, a common sample is chosen as an example here, where the splat generated by the droplet mentioned before with diameter of $4.59\,\mu m$ is impacted by a droplet with diameter of $6.8\,\mu m$ at a velocity of $231\,m/s$ and temperature of $3795\,K$. The point of impact for this droplet is nearly $14\,\mu m$ away from the center of impact of the first droplet. Result of this binary impact is shown in figure 5.15. As the time between the two impacts is much longer than the solidification time, it is safe to assume that the first droplet’s splat has already solidified by the time the second droplet impacts. It can be observed that the second droplet becomes more spread in the free directions where there were no obstacles, in contrast to the direction where the splat from first droplet is present. In that direction, the second splat has spread into three main branches, two bypassing the first droplet’s solidified splat, and one being forced to splash over it.
Figure 5.13: Heat flux at the impact point of the droplet on the substrate as a function of non-dimensional time ($t^* = tV/D_0$), shown near the time of impact (figure on top) and afterwards (figure on bottom).
Figure 5.14: Solidified splat from a droplet with 4.59µm diameter, impacting the substrate at 175ms with a temperature of 3517K, shown at $t^* = 1500$. 
Figure 5.15: Binary impact of two droplets with diameters of 4.59 and 6.8µm at velocities of 175 and 231 m/s and temperature of 3517 and 3795K. The position of impact is offset by 14µm.
5.5 Predictions for Finished Coating

Equation 5.1 is now used in a Matlab subroutine to examine the droplets collected on the substrate from SPS results. Having an approximation for spread of each droplet upon impact allows us to calculate the number of splats that will overlap in forming the final coated substrate. Results obtained are shown in table 5.3. It can be seen that near 22\% of the total droplets hitting the substrate will generate splats with binary overlaps. In addition, 10\%, 5\%, and 2\% will have two, three, and four splat overlappings, respectively. The rest of overlaps start to fall below 1\% of the total droplets. The maximum number of overlaps is 12 with only 3 occurrences here. Overall, the finished coating of the substrate will be dominated by 60\% isolated impacts and 22\% binary-overlapped impacts. Finally another subject of interest is efficiency calculation for the total spray process. For this calculation, the total area of the substrate that is covered by solidified splats is needed. Using the correlation derived here for the final splat sizes (equation 5.1), the total covered area can be calculated. Overlaps of different plats however needs to be subtracted from calculated area. As multiple overlaps are occurring here, the calculation is conducted in another manner. The substrate surface is meshed in fine resolution. The summation of the areas of those cells that fall under at least one splat will result in the total area of substrate that has been covered. The resolution of this mesh has been increased till no noticeable variation in calculated area is observed. Resulted total area of the substrate that has been successfully coated is 80.57 mm$^2$. The total area of this part of the substrate has also a value of 4975 mm$^2$.

By applying the information gathered so far to cases A to E, effects of different parameters of the plasma torch on the finished surface can be studied. Taking droplets’ spread according to equation 5.1 and assuming no pore formations occurring between
the splats, the final coated surface for cases A to E can be predicted as shown in figure 5.16. The results show that after coating, the highest points from the substrate’s surface are 4.28, 1.07, 2.44, 5.28, and 1.65 μm for cases A to E, respectively.

It has to be noted that by considering the temperature particles and omitting droplets that have not fully melted, the substrate will be clear for cases A, B, and C. These are droplets that have not received enough energy from the torch to finished their evaporation and melting cycles completely. Under this condition, the substrate for cases D and E is shown in figure 5.17. The maximum height of the coating on the substrate is 2.14 and 1.65 μm for cases D and E, respectively. The coating does not change significantly for case E, as most of the particles under these torch conditions reach the substrate being fully molten. Results for case E show that the points on the positive side of the x axis have higher heights compared to the left side. This is due to the injection angle for the suspension stream, which points from positive x values towards negative values (right to left).

5.6 Pore Formation

A two-dimensional setup has been used here to study the cases where droplet impacts are prone to void formation. For the test cases below, a droplet with a radius of 3 μm is impacted on a surface. The surface geometry is changed going from a flat surface to surfaces with step sizes of 0.12, 0.24, 0.36, 0.54 μm. Impact velocity is 150 m/s for all the cases. Initial temperature of the substrate is 300 K. All figures have been reported at \( t = 0.1 \mu s \), which is when the spread and recoil of the droplet have already finished and the shape of the splat is not going through any significant changes.

In the first case, the initial temperature for the droplet is 2985 K. This temperature is slightly above solidification point of YSZ. Results for this test have been shown in
Figure 5.16: Finished surface of the substrate for cases A to E, colored by the height from the substrate’s surface.
Figure 5.17: Finished surface of the substrate for cases D and E taking particle temperatures into account, colored by the height from the substrate’s surface.

Since the drops here are close to their melting point, solidification occurs shortly after they come in contact with the substrate. This prevents any significant pore formation near the step dent on the surface. However, as test cases described in table 5.2 show, with the right choice of parameters for the torch and injection, drops reaching the substrate are at higher temperatures, mostly way above the melting point of YSZ. Hence, the same test case is repeated here with now an initial temperature of $3500K$ for the droplets. Results for this case have been shown in figure 5.19. In these cases, as the droplet needs more time to solidify, the spread and recoil occurs while the droplet has not lost enough heat for solidification. The impact this time is being controlled by the momentum force spreading the drop over the step boundary and surface tension forces acting upon it. Here the pore that is formed is nearly a quarter of circle whose radius is very close to the step height. A possible explanation for this behaviour might be due to large surface tension magnitude of molten YSZ, which forces the splat to form a nice curvature over the discontinuity. It is evident where the temperature was close to melting point of YSZ, the solidification process
has prevented pore formation.

Figure 5.18: Normal impact of droplet with a radius of 3µm on flat surface against surfaces with step sizes of 0, 0.12, 0.24, 0.36, and 0.54µm, from top to bottom respectively. Impact velocity is 150m/s and the initial temperature for the droplet and the substrate are 2985 and 300K. Current time for all cases is 0.1µs.

An important case where gaps and voids may appear is during the coating process
Figure 5.19: Normal impact of droplet with a radius of $3\mu m$ on flat surface against surfaces with step sizes of 0, 0.12, 0.24, 0.36, and $0.54\mu m$, from top to bottom respectively. Impact velocity is $150m/s$ and the initial temperature for the droplet and the substrate are 3500 and $300K$. Current time for all cases is $0.1\mu s$. 
of a rough surface. Several researches like the works of Xue et al. [121], Shinoda et al. [122], and Parizi et al. [123] have focused on this topic, using a patterned substrate making conditions better for studying. In these studies, YSZ droplets are impacting patterned solid surfaces. The size of these droplets, however, are typically large, in the orders near 15\(\mu m\). As this size is large compared to SPS-generated particles, an alternative test is used. Here, seven droplets with a size of 3\(\mu m\) have been used on the same substrate.

Before seeing results for this case, for comparison reasons, initially two test cases similar to previous studies are conducted here. In the first, similar to Xue et al. [121], a drop with diameter of 18\(\mu m\) is impacting a patterned solid surface at a velocity of 250\(m/s\) and temperature of 3173\(K\). The surface is patterned with columns with a width of 4\(\mu m\), height of 3.3\(\mu m\), and distance of 1\(\mu m\). Schematic drawing for this test is shown in figure 5.20. Results of this simulation are shown in figures 5.21 to 5.23. The impact point here has been located over the middle solid column. In the second case, similar to Shinoda et al. [122], the same test is repeated, this time with a diameter of 15\(\mu m\) and the impact point for the droplet is located in the middle of a column void. Results for this simulation are shown in figures 5.24 and 5.25. As it can be seen, similar to reports of Shinoda et al., results here show that the YSZ impact will fill 5 voids in the middle of the substrate and get scattered over the rest. Closeup of the three middle voids showing the movement of the solidification front is shown in figure 5.26. The material in middle column, being in contact with the substrate the longest, has started to solidify first. By near \(t^* = 250\), all the material inside the three voids has completely solidified. The rest of the material solidifies here nearly by \(t^* = 1000\).

Void formations inside each column have been compared here against experimental and analytical results of Xue et al. [121]. This comparison is shown in figure 5.27.
Figure 5.20: Two-dimensional simulation of droplet with diameter of 18 $\mu m$ impacting a patterned surface at velocity of 250 m/s.
Figure 5.21: Droplet with a diameter of 18\(\mu\)m impacting a patterned substrate at velocity of 250\(m/s\) and temperature of 3173\(K\).
Figure 5.22: Closeup at $t^* = 0.5$ for a droplet with diameter of 18µm impacting a patterned substrate at velocity of 250m/s and temperature of 3173K.
Figure 5.23: Heat flux magnitude (W/m²) for a droplet with diameter of 18µm spreading on a patterned substrate at velocity of 250m/s and temperature of 3173K at nearly $t^* = 0.6$. 
Figure 5.24: Droplet with a diameter of 15\(\mu\)m impacting a patterned substrate at velocity of 250\(m/s\) and temperature of 3173\(K\).
Figure 5.25: Droplet with a diameter of 15\(\mu\)m impacting a patterned substrate at velocity of 250\(m/s\) and temperature of 3173\(K\).
Figure 5.26: Closeup of the solidification front movement inside the column voids, after impact of a droplet with diameter of 15\(\mu m\) on a patterned substrate at velocity of 250\(m/s\) and temperature of 3173\(K\).
The two dots at each distance are for the columns to the left and right of the impact point. Results indicate that for voids close to the impact point, the pressure under the drop will force a complete filing of the columns. As the splat spreads away, the pressure is lost and the voids are filled with the fluid being forced into the patterns from the sides, and columns are filled with a circulating flow pouring in them. As current simulation is single phase, effects of air entrapment would not be captured. The test case here is also performed in two dimensions, which can be a source for the differences for columns located further from impact point.

Figure 5.27: Void formation as a function of distance from impact point, against experimental and analytical results of Xue et al. [121].

As the size of droplets in suspension plasma spraying is in order of a couple of micrometers, these tests are repeated here using droplets with smaller sizes, as shown in 5.28. As droplets reaching the substrate can have different temperatures, it has also been assumed here that the droplets will reach the substrate at different temperatures above melting point of YSZ. While in theory if the SPS injection is ideal,
as shown before, most particles will reach the substrate at temperatures much higher than melting point, in practice, particles will not be all at a high quality. Hence, the temperatures for the seven droplets used in the following test, shown in figure 5.28, are chosen to be 2985, 3000, 3200, 3400, 3500, 3000, and 3000K, for droplets #1 to #7, respectively. The material for the droplets is YSZ. The substrate has a thermal conductivity of Tin, i.e. $66W/(m \cdot K)$. For simplicity, the rest of physical properties of the substrate are identical to the droplets.

Results for this test at various values of $t^*$ have been plotted in figures 5.29 to 5.33. Findings suggest that since the size of the droplets is comparable to the size of patterns, the splat reaching the void first will most likely contribute to filling the void. Also for a void to be filled with more matter, the impact site needs to be closer to its center. For instance, the void to the left of droplet #3 is only partially filled compared to the rest, as the passive impact of this droplet with respect to this void does not provide the splat with a downward spread direction. Binary collision of spreading splats, as of droplet #1 and #2, can provide the downward momentum direction needed for filling the void. The voids with most penetrated matter here are a byproduct of direct impacts of the droplets, as seen for droplets #4 and #5. Here, droplet #1 spreads over the void to its right. By the time drop #4 reaches this void, the splat of #1 has already covered the opening. As the splat has not yet solidified, droplet #4 pushes in what is already on the top of this void and fills the rest by its own, which can be seen in figure 5.30. The final filling of this void has come from #1 at the bottom, and #4 in the heart of filling, clearly identified from drop’s #1 and #4 initial temperature differences. The temperature of the fillings in voids will be closer to the drop that reaches the void sooner. It has to be noted that all these are under the assumption that impacts are happening shortly one after another, with small time for solidification to occur. If the impacts occur over a longer period of
time, there would possibly exist many column openings that are covered by solidified splats. Moreover, effects of air entrapment are neglected here, as the gas phase is not simulated. With the assumption of having impact shortly one after another, final droplets will hit the molten bath formed below them. In this particular test case here, droplets #5-#7 do not come in direct contact with the substrate. Impact of droplets #5 and #6, being close, cancels the inward splashes creating a calm zone in the middle of the substrate. Droplet #7, hits this center, causing a washout where the multiple outward splashes are formed that will carry considerable amount of molten matter away from the impact site.

Heat flux values between the substrate and droplets and also among the droplets at $t^* = 1.9$ is shown in figure 5.34. Magnitude of velocity and acceleration components due to pressure, viscous, and surface tension forces are shown in figure 5.35 at $t^* = 1.3$. It becomes evident here that as expected, the effects of surface tension forces are negligible at this stage of simulation compared to pressure and viscous effects. The final material that becomes frozen and fills the void between the columns has been shown in figure 5.36. The solidification process is completely finished by $t^* = 15000$, which is higher than superposition of 7 droplets on a flat surface ($t^* \approx 1000 - 1500$ for each droplet), even though the substrate surface is expanded due to existence of patterns compared to a flat surface. This can be explained by noting the fact that most of the molten matter has not completely penetrated the voids leaving gaps behind. Hence, the liquid materials are accumulated on top of one another, decreasing the effective area of the substrate that comes in direct contact, leading to lower heat transfer rates to the substrate.

The main assumption in the aforementioned example is having the molten droplets impacting with short time intervals between them. SPS results, however, suggest this will be a rare occurrence. Hence it is reasonable to repeat the same test case here
Figure 5.28: Impact of seven droplets on a patterned surface at 150 m/s, colored by temperature (K).

Figure 5.29: Coating process of a patterned substrate at $t^* = 1.3$, colored by temperature (K).
Figure 5.30: Coating process of a patterned substrate at $t^* = 2.5$, colored by temperature ($K$).

Figure 5.31: Coating process of a patterned substrate at $t^* = 3.1$, colored by temperature ($K$).
Figure 5.32: Coating process of a patterned substrate at $t^* = 7.3$, colored by temperature ($K$).

Figure 5.33: Coating process of a patterned substrate at $t^* = 11.5$, colored by temperature ($K$).
with an improved condition. In the following test case, it has been assumed that each of the 7 droplets will impact individually and solidify. Only then the next droplet impact is executed. Result for this simulation is presented in figure 5.37. It can be seen that unlike previous cases, the chaotic behavior of splashes are now limited. The largest void penetration this time has been obtained from impact of droplet #3. Both droplets #1 and #3 possess passive impacts with respect to the voids next to them. Droplet #3, however, being nearly 200 K hotter than #1, has had more time to spread before the solidified layer underneath immobilized it. Final coated splat shows two large voids being formed between the layers. The lower void, between droplet #1 and #4 and the upper void is surrounded by #5, #6, and #7. In the lower void, the initial gap underneath the splat of droplet #1 has a height of 0.25 μm, while the gap underneath droplet #6 splat in the upper void measures at nearly 0.5 μm, which can explain the size difference between these two voids. The area of these voids are $2.5 \times 10^{-13}$ and $1.2 \times 10^{-13} m^2$ for upper and lower voids, respectively. Moreover, two
Figure 5.35: Magnitude of velocity (V) and acceleration components due to pressure (p), viscous (v), and surface tension (s) forces at $t^* = 1.3$. 
smaller voids between the splats and substrate exist with areas of $9.6 \times 10^{-14}$ and $6.1 \times 10^{-14} m^2$. There are also 32 smaller voids between the splats of the 7 droplets with an approximate average area of $4.7 \times 10^{-15} m^2$ per void.

Figure 5.36: Solidified material left in the voids at $t^* = 15000$.

Figure 5.37: Solidified material on the substrate with the assumption of having individual impacts and solidifications for each of the 7 droplets.
Using the information on void formations, it is useful to repeat the test case on prediction of final coated surface using Matlab routine and account for the void formations. To save computational time, only test case E has been chosen for further study here. For enhancing the predictions, more SPS droplets for case E have been obtained from the numerical simulation, bringing the total number of droplets gathered on the substrate to near a million, sprayed over 0.61s. Equation 5.1 has been used for predicting the spread diameter of SPS particles on the substrate. It has been assumed that molten particles form discs on the substrate. The tangential component of the impact velocities has been neglected here. Results of simulations in previous sections suggested that when a droplet at high temperature impacts an uneven surface, there is a high chance for void formation with a similar size to the height of the void. This has also been included here by making the assumption that droplets impacting the surface with high temperatures will contribute to formation of voids beneath them, although the dimensions of the voids are small compared to other dimensions. Application of these procedures to the example of five droplets of table 5.1 is shown in figure 5.38. It can be observed that using this method, the overall behavior of impacts can be predicted as the high and low points in both simulation and prediction are close. Visualization of the substrate’s surface for the deposition of this 1 million droplets has been shown in figure 5.39. Results indicate that over the 0.61(s) spray time, 69.80% of the total solid mass of YSZ initially injected into the domain has been deposited onto the substrate, leading to a deposition efficiency of 69.80%. Growth of the area of the solidified material on the substrate as a function of time for this 1 million droplets has been shown in figure 5.40.
5.7 Conclusion

Results obtained from previous studies using Finite Volume solution to Suspension Plasma Spraying was closely examined using a Smoothed Particle Hydrodynamics (SPH) solver. It is shown that due to mist-like fragmentation of the liquid suspension, the substrate is coated gradually over a large area with sub-micron particles. Combining the wide range of YSZ droplets collected on the substrate, with the splat formation and solidification results obtained from the SPH solution, shows that the surface is coated by isolated particles that will hit the substrate and solidify far from each other. The droplets studied here will mostly have completed their impact and recoil cycle by $t^*$ values below 10. For the splat to fully cool down and solidify, it has to remain on the substrate for $t^*$ values near 1000 to 1500. This is mainly due to the high temperature of the molten ceramics at impact time. Comparing these results with findings from Finite Volume solution confirms that droplets will solidify before being hit by another splat (wait time between impacts is $t^* = 300,000$ to 5
Figure 5.39: Solidified material on the substrate for 1 million droplets obtained from numerical superposition of splats on the substrate (colored by height from the surface of the substrate).
Figure 5.40: Growth of the area of the solidified material on the substrate as a function of time for 1 million droplets obtained from numerical superposition of splats on the substrate.

million), hence, it is very rare for two droplets to hit one another on the substrate while both being in a molten state. Findings also suggest noticeable differences between the first coating layer compared to others. For YSZ droplets that come in direct contact with metallic substrates, higher thermal conductivity of the substrate material will contribute to fast formation of a solid layer beneath the splat that will eventually immobilize it. For next layers of coating, where YSZ droplets hit solidified YSZ splats, lower thermal conductivities of the solid matter will delay the cooling process, resulting in a more free spread and splash of YSZ droplets on the substrate.
Chapter 6

Closure

6.1 Conclusions

Suspension Plasma Spraying is an emerging method for improving quality of coatings. While different studies have been conducted on various stages of this process, a comprehensive study is missing to collect and combine different available methods into one solution. This thesis, then, presented comprehensive study of the process using various methodologies for enhancement of numerical results. Different procedures are used for a better study of the phenomena involved, from injection port to the solidification of splats on the substrate.

Injected suspension droplets are tracked during flight using a coupled Finite Volume-Discrete Phase solver. Change of properties for droplets during flight is taken into account. Results clearly indicate utilizing a proper viscosity model for the suspension in droplets, as it goes through large concentration changes, is vital in capturing accurate flight paths and breakup patterns.

Moreover, the effect of different parameters on final deposition of particles and flight conditions is evaluated. Injection position, angle, and velocity, along with torch
operating conditions such as flow rate and power are varied to create a range of test cases for better understanding of the suspension spraying process. These cases show that the final fate of the injection particles can be controlled by changing the parameters involved.

Different parameters, however, have different impacts on the overall outcome. For injector operating conditions, test cases show that injection of suspension at high mass flow rates can result in a complete penetration through the plasma plume which will lead to near to non deposition. It can also be concluded that under the conditions of the current study, the best SPS deposition rates are achieved when injection needle is placed close to the torch centerline and near 1cm from nozzle exit. Droplets also need to be injected at a low velocity. These injection conditions introduce the least amount of disturbances in torch flow patterns and allows more droplets to be carried towards the substrate. For injection of water suspensions, it is clearly important to have enough power at the torch to make sure the cooling effects of evaporating water is recovered. Otherwise in many cases, the liquid content might not effectively evaporate and lead to the impact of wet mixtures on the substrate. Results here also indicate that increasing the torch power enhances particle qualities on the substrate better compared to increasing the inlet mass flow rate for the torch.

Modeling heat transfer with phase change in SPH has also been investigated as a tool for capturing impact and solidification of suspension droplets on the surface. The release/absorption of latent heat during phase change is accounted for by modifying either the heat capacity in the energy equation or by adding a source term.

For effective heat capacity method, a new approach is introduced, which uses smoothing and superposition of two kernels to gradually release/absorb the latent heat near the phase change temperature. Compared to a number of alternatives, this new approach yields accurate solutions while limiting the computational cost. This model,
however, heavily relies on the choice of the smoothing kernel, and using it without proper modification for free surface flows will be problematic due to neighborhood interruptions near the surface. The enthalpy method, on the other hand, while being computationally more expensive, provides a good platform for free surface flows that are of interest in current study. It also provides the opportunity of modeling pure molten materials without the need of a phase change interval. Last but not least, it has to be mentioned that the SPH approximations proposed here, although based on a SPH fundamentals, are also applicable to grid-based methods.

Finally, connecting the FV and SPH solver for the case of Suspension Plasma Spraying revealed statistical information on droplet depositions on the substrate. It is shown that due to mist-like fragmentation of the liquid suspension, the substrate is coated gradually over a large area with sub-micron particles. Combining the wide range of YSZ droplets collected on the substrate, with the splat formation and solidification results obtained from the SPH solution, shows that the surface is coated by isolated particles that will hit the substrate and solidify far from each other. The droplets studied here will mostly have completed their impact and recoil cycle by \( t^* \) values below 10. For the splat to fully cool down and solidified, it has to sit on the substrate for \( t^* \) to grow to values near 1000 to 1500. This is mainly due to the high temperature of the molten ceramics at impact time. Comparing these results with findings from FV solution confirms that droplets will solidify way before being hit by another splat (\( t^* \) wait time between 300,000 to 5 million), hence, it is very rare for two droplets to hit one another on the substrate while both being in a molten state.

To summarize, in this thesis,

- a numerical solution was implemented to calculate the trajectory, breakup, evaporation, and melting of suspension droplets from injected till reaching the sub-
strate.

- effects of increase of concentration with evaporation of solvent on thermal and physical properties of suspension were included in the calculations, especially for the viscosity of the droplets.

- effects of plasma jet on solid particles, such as Knudsen effect and high temperature gradients, were taken into account.

- different operating conditions, such as torch and injection parameters, were studied for better understanding of their effects on the quality of droplets impacting the substrate.

- numerical models for handling phase change were developed using Smoothed Particle Hydrodynamics (SPH) and by inclusion of latent heat release (absorption) during solidification (melting) into effective heat capacity and enthalpy formulation.

- numerical models were validated against various available analytical, experimental, and numerical results in literature.

- the model was applied for predicting the quality of a SPS coating over a brief spray time window.

### 6.2 Recommendations for future Work

These topics are recommended for making improvements to the current research:

- During solidification, as the density of the forming solid matter is usually larger than the liquid, the material shrinks to provide more mass per volume over the
solidified portion. Further studies are needed in robust and stable implementation of the shrinkage process. The amount of change in volume and density due to shrinkage can be quantified by defining a shrinkage factor as

\[ S.F. = \frac{\rho_s - \rho_l}{\rho_l} \]  

(6.1)

During shrinkage, the density values at solidified portions needs to become larger than liquid state. Taking local mass to remain constant, shrinkage leads to a decrease in local volume at solidified points. To numerically capture this, it can be assume that all SPH particles are spheres with a radius of \( k_h \). Hence, the volume of each particle becomes \( V = \frac{4}{3}\pi(k_h)^3 \). This means the density of each particle is roughly proportional to \( \frac{1}{k^3} \). Substituting this into equation 6.1 gives

\[ S.F. = \frac{h_s^3 - h_l^3}{h_l^3} \]  

(6.2)

Taking \( S.F. \) to be known for any material of interest through experimental data, particle’s radius after solidification becomes

\[ h_s = \frac{1}{\sqrt{S.F. + 1}} h_l \]  

(6.3)

Taking \( \beta \) to be the solid fraction on each particle, the radius of each particle during solidification using linear interpolation becomes

\[ h = h_l + \beta(h_s - h_l) \]  

(6.4)

For applying shrinkage effects on the radius of particles, another procedure can
also be used. The density of each particle can be calculated using

\[ \rho = m \sum_j W(x_{ij}, h) \]  

Substituting this into equation 6.1 gives

\[ S.F. = \frac{m \sum W(x_{ij}, h_s) - m \sum W(x_{ij}, h_l)}{m \sum W(x_{ij}, h_l)} \]  

Assuming mass of each particle remains constant during phase change, which also guarantees global conservation of mass, leads to

\[ S.F. = \frac{\sum W(x_{ij}, h_s) - \sum W(x_{ij}, h_l)}{\sum W(x_{ij}, h_l)} \]  

The value of \( h_s \) can be numerically calculated at the beginning of the simulation by solving equation 6.7 for a known shrinkage factor. This value can then be used in equation 6.4 to update radius of particles during simulation where phase change is occurring.

Pressure in SPH is typically calculated from the density variations using formulations like

\[ p = p_0 \left[ \left( \frac{\rho}{\rho_0} \right)^\gamma - 1 \right] \]  

As is evident, the increase of density values during shrinkage process can leave a negative impact on the calculated pressure. During solidification, density becomes larger than \( \rho_0 \), which generates a pressure wave from shrank areas towards the liquid phase. Although the solidification process along with mushy zone implementations dampens some of this effect, it can still affect computa-
tional results. A path to a suitable remedy might be by modifying equation 6.8 in the form of

\[ p = p_0 \left[ \left( \frac{\alpha \rho}{\rho_0} \right) \gamma - 1 \right] \]  

(6.9)

In equation 6.9, only the liquid portion of each particle contributes to pressure calculations.

The formulation mentioned above still fails in practice to create a stable and predictable implementation of shrinkage. Variations in pressure are the main source of instabilities that would lead to unrealistic density calculations post shrinkage. Further studies are needed for better handling of this phenomena.

- Stability issues for some SPH formulations is a known concept. SPH formulations, specially in classical form could become unstable if the solution is allowed to run for long time periods. The formulations used in this work are no exception. Various numerical methods have been proposed by others for addressing these issues, many of which have led to development of new SPH platforms, such as XSPH [124] and ISPH [125]. Obtaining true incompressibility is the main goal. Further studies can be performed on migration of these tools to current framework.

- The surface tension method used in current work, though being widely popular, has some shortfalls. For instance, the value of surface tension coefficient is not an input and hence calibration is needed. Furthermore, relying on cancellation of surface tension forces between particles can effectively make the solution resolution dependent. More robust surface tension models that use continuum surface forces (CSF) are available but mostly are not applicable to free surface
SPH and are computationally expensive, since the liquid interface needs to be reconstructed before obtaining curvature values.

- Thermal and mechanical stresses that form from impact and solidification of droplets are neglected in current framework. A study with inclusion of solid-liquid effects can be found at [29].

- The position of the solidification front calculated in the numerical results here has been used as a validation tool, similar to many other numerical studies. There are however reports on shortfalls of this comparison tool and alternative procedures have been suggested [21].

- Thermal contact resistance between the substrate and the drop has been neglected here. Inclusion of this will enhance accuracy in capturing the actual physical phenomena [1].

- The value of contact angle in current framework is assumed to remain constant during impact and spread. It is however more reasonable to use a dynamic contact angle model, similar to [58].


[37] E. Meillot, S. Vincent, C. Caruyer, D. Damiani, and J. Caltagirone. Mod- 
elling the interactions between a thermal plasma flow and a continuous liquid 

[38] K. Toda and H. Furuse. Extension of Einstein’s viscosity equation to that for 
concentrated dispersions of solutes and particles. *Journal of Bioscience and 

formation and coating microstructure using the solution precursor plasma spray 


model for miscible flow in three-dimensional fractures and the two-dimensional


