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A Computational Approach for Modeling Plasma Sprayed Coatings from Splat to Bulk Deposits

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FLORIDA INTERNATIONAL UNIVERSITY

Miami, Florida

A COMPUTATIONAL APPROACH FOR MODELING PLASMA SPRAYED
COATINGS FROM SPLAT TO BULK DEPOSITS

A thesis submitted in partial fulfillment of

the requirements for the degree of

MASTER OF SCIENCE

in

MECHANICAL ENGINEERING

by

Sadhana Bhusal

2019

To: Dean John L. Volakis
College of Engineering and Computing

This thesis, written by Sadhana Bhusal, and entitled A Computational Approach for Modeling Plasma Sprayed Coatings from Splat to Bulk Deposits, having been approved in respect to style and intellectual content, is referred to you for judgment.

We have read this thesis and recommend that it be approved.

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Florida International University, 2019

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DEDICATION

I dedicate this thesis to my family and friends for always believing in me and inspiring me to follow my dreams and loving me unconditionally.

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Whatever I am today, I owe it all to you.

ABSTRACT OF THE THESIS

A COMPUTATIONAL APPROACH FOR MODELING PLASMA SPRAYED
COATINGS FROM SPLAT TO BULK DEPOSITS

by

Sadhana Bhusal

Florida International University, 2019

Miami, Florida

Professor Arvind Agarwal, Co-Major Professor

Professor Benjamin Boesl, Co-Major Professor

In this study, a multi-pronged computational approach is developed to predict the effect of spray parameters on aluminum oxide splat formation and mechanical properties of the coatings. The splat morphology is investigated using computational fluid dynamics approach. Simulated splat morphologies show a good agreement with the experimentally obtained splats. Three-dimensional coating structure is constructed using the stochastic approach using simulated splat morphologies. Finite Element Analysis is used to compute the elastic modulus of the coating. An inter-splat correction factor is introduced which considers inter-splat cracks, interface bonding and other effects like curling of splats and splat sliding. After the correction factor, the computed elastic modulus for simulated coating is comparable to the experimental values (4~5%). This study shows that the proposed computational approach can predict the mechanical properties of the coating and is promising for developing plasma-sprayed coatings with predictable properties and can be extended to other materials systems.

TABLE OF CONTENTS

CHAPTER	PAGE
CHAPTER 1. INTRODUCTION.....	1
1.1 Plasma Spray	1
1.2 Splats.....	2
1.3 Computational Methods and Their Limitations.....	3
1.4 Research Objective	4
CHAPTER 2. LITERATURE REVIEW.....	8
2.1 Processing Parameters in Plasma Spray	8
2.2 Experimental Techniques	11
2.3 Computational Techniques	13
CHAPTER 3. MATERIALS AND METHODS.....	18
3.1 Experimental Methods.....	18
3.1.1 Processing.....	18
3.1.1.1 Powder.....	18
3.1.1.2 Substrate	18
3.1.1.3 Processing Parameters	19
3.1.1.4 Formation of Splats and Coatings	20
3.1.1.5 In-flight Particle Diagnostic Sensor	21
3.1.2 Characterization.....	22
3.1.2.1 Optical Microscopy	22
3.1.2.2 Porosity Measurement.....	22
3.1.2.3 Nanoindentation	22
3.2 Computational Methods.....	23
3.2.1 Simulation of Single Splats Using Computational Fluid Dynamics Approach.....	23
3.2.2 Formation of Three-dimensional Coating Structure Using Stochastic Models.....	28
3.2.3 Object-Oriented Finite Element Analysis for Elastic Modulus Prediction	31
CHAPTER 4. RESULTS AND DISCUSSION.....	33
4.1 Formation and Simulation of Splats	33
4.2 Simulated and Experimentally Obtained Coating Microstructure.....	43
4.3 Computed and Experimental Elastic Modulus of the Coating	47
CHAPTER 5. CONCLUSIONS	52
CHAPTER 6. RECOMMENDATIONS FOR FUTURE WORK	53
LIST OF REFERENCES	57

APPENDIX.....	65
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LIST OF TABLES

TABLE	PAGE
Table 3-1. Processing parameters used in the formation of splats and coatings.....	19
Table 3-2. Thermo-physical properties of Aluminum Oxide for splat simulation [77]	26
Table 3-3. Particle velocities and temperatures for different plasma power obtained from inflight diagnostic sensor during splat formation.....	26
Table 3-4. Smallest distance (l) between two neighboring splats based on the equations describing splat shapes (Based on the stochastic model developed by Ghafouri-Azar et al. [64]	30
Table 4-1. Elastic modulus obtained from nanoindentation and OOF2 for the cross- section	49
Table 4-2. Elastic modulus obtained from nanoindentation and OOF2 with an inter- splat correction factor for the cross-section	51
APPENDIX Table 1. Parameters used for formation of 2D image for Power 28 kW and substrate temperature 100 °C	65
APPENDIX Table 2. Parameters used for formation of 2D image for Power 28 kW and substrate temperature 180 °C	67
APPENDIX Table 3. Parameters used for formation of 2D image for Power 28 kW and substrate temperature 250 °C	69
APPENDIX Table 4. Parameters used for formation of 2D image for Power 32 kW and substrate temperature 100 °C	71
APPENDIX Table 5. Parameters used for formation of 2D image for Power 32 kW and substrate temperature 180 °C	73
APPENDIX Table 6. Parameters used for formation of 2D image for Power 32 kW and substrate temperature 250 °C	75
APPENDIX Table 7. Parameters used for formation of 2D image for Power 35 kW and substrate temperature 100 °C	77
APPENDIX Table 8. Parameters used for formation of 2D image for Power 35 kW and substrate temperature 180 °C	79

APPENDIX Table 9. Parameters used for formation of 2D image for Power 35 kW and substrate temperature 250 °C	81
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LIST OF FIGURES

FIGURE	PAGE
Figure 1-1. Schematic Diagram of Plasma Spray	1
Figure 1-2. Morphologies of splats	2
Figure 1-3. Schematic Diagram of Computational Approach	5
Figure 2-1. Nickel splat morphologies on AISI 304 substrate. (a) Splash splat, $T_s = 300$ K. (b) Disk splat $T_s = 673$ K. (c) Fraction change of disk splat with substrate temperature [40]	9
Figure 2-2. Morphologies and 3-D surface profiles of zirconia splats prepared on (a) and (c) cold substrate ($100\text{ }^{\circ}\text{C}$) and (b) and (d) hot substrate ($250 - 300\text{ }^{\circ}\text{C}$) [19].....	12
Figure 3-1. Schematic Diagram for Plasma Spray Setup	20
Figure 3-2. Operational Principle of AccuraSpray Diagnostic Sensor [67]	21
Figure 3-3. Maximum spread ratio for different values of CPR.....	28
Figure 4-1. Comparison of splat morphology obtained from experiments and simulations, a) Power 28 kW, Substrate Preheat Temperature $100\text{ }^{\circ}\text{C}$, b) Power 28 kW, Substrate Preheat Temperature $180\text{ }^{\circ}\text{C}$ and c) Power 28 kW, Substrate Preheat Temperature $250\text{ }^{\circ}\text{C}$	34
Figure 4-2. Comparison of splat morphology obtained from experiments and simulations, a) Power 32 kW, Substrate Preheat Temperature $100\text{ }^{\circ}\text{C}$, b) Power 32 kW, Substrate Preheat Temperature $180\text{ }^{\circ}\text{C}$ and c) Power 32 kW, Substrate Preheat Temperature $250\text{ }^{\circ}\text{C}$	35
Figure 4-3. Comparison of splat morphology obtained from experiments and simulations, a) Power 32 kW, Substrate Preheat Temperature $100\text{ }^{\circ}\text{C}$, b) Power 32 kW, Substrate Preheat Temperature $180\text{ }^{\circ}\text{C}$ and c) Power 32 kW, Substrate Preheat Temperature $250\text{ }^{\circ}\text{C}$	36
Figure 4-4. Volume fraction of splats for a) 28 kW, b) 32 kW and c) 35 kW plasma power	38
Figure 4-5. Splat size distribution for different substrate preheat temperatures for a) 28 kW, b) 32 kW and c) 35 kW plasma power	40
Figure 4-6. Average diameter of different splat morphologies for a) 28 kW, b) 32 kW and c) 35 kW plasma power	41

Figure 4-7. Formation of three-dimensional coating structure from simulated splats.....	43
Figure 4-8. Cross-sectional image of coating, a) from experiments power 32 kW, substrate preheat temperature 180 °C and b) from computational stacking power 32 kW, substrate preheat temperature 180 °C.	44
Figure 4-9. Deposition of real time plasma process and computational model	45
Figure 4-10. Comparison of the porosity of the cross-sectional orientation of the coatings obtained from finite element analysis (FEA) for the computational coating structure and ImageJ for the experimental coatings	46
Figure 4-11. Finite element analysis, acquiring an image, adaptive meshing, and application of boundary conditions.....	48

LIST OF ABBREVIATIONS

Symbol	Description
HVOF	High Velocity Oxygen Fuel
VOF	Volume of Fluid
V	Velocity Vector
P	Pressure
ν	Kinematic viscosity
ρ	Density
F	Body forces
h	Enthalpy
k	Thermal conductivity
T	Temperature
f	The fraction of a cell volume occupied by liquid
∇	Vector differential operator
R	The radius of the impacting droplet
D_n	The diameter of the impacting droplet
l	The distance between the droplet impact point and the center point of previously deposited splats
a	Major radius of splat (measured along Y-axis)
b	Minor radius of splat (measured along X-axis)

CHAPTER 1. INTRODUCTION

1.1 Plasma Spray

Plasma Spray is one of the thermal spray techniques in which a DC arc is generated between the anode and cathode resulting in the dissociation and ionization of gases to form plasma [1]. The temperature of plasma can reach up to 10,000 K making it suitable for the formation of metallic, ceramic as well as polymeric coatings [2]. The material in the form of powder is injected in the plasma stream where it is rapidly heated and accelerated to a high velocity towards the substrate material [3–5]. The coating is formed by the buildup of the successive layers of molten particles which flatten and solidify to form a lamellar microstructure. Due to the high cooling rates typically of the order of 10^6 to 10^8 K/sec, the microstructures of the coatings are fine-grained and homogeneous [6,7]. The schematic diagram of the plasma spray process is shown in Figure 1-1.

Figure 1-1. Schematic Diagram of Plasma Spray

Plasma sprayed coatings are widely used due to their flexibility (any material can be deposited on any substrate), ability to produce homogeneous coating structure, the formation of thick coating structure and ability to produce coating structure in different environments (air, inert gas, vacuum) [8]. Plasma sprayed coatings find a wide range of applications in the automotive, aerospace, chemical, textile and biomedical industries [9]. The typical applications of plasma sprayed coatings are wear and erosion resistance for various machinery parts, improvement of friction properties, the formation of electrodes, biomedical coatings for the orthopedic and dental prosthesis, thermal and chemical barrier coatings for the piston, gas turbine blades, corrosion protection of equipment, etc. [10–12].

1.2 Splats

During the deposition and solidification of the molten particle, a lamellar structure called “splat” is formed [13]. The pancake-shaped splats are the building blocks of the plasma sprayed coating [14]. The thermal, mechanical and electrical properties of plasma sprayed coatings are dependent on the morphology, and arrangement of the splat [15–17]. There are three major shapes of splats commonly observed in the plasma sprayed coatings- disk, fragmented and fingered (splashed) splats [18] as shown in Figure 1-2.

Figure 1-2. Morphologies of splats

Disk-shaped splats have better contact with substrate resulting in the increased adhesion and reduced porosity [19]. Presence of the fragmented and splashed splats in the coating structure increases porosity and pose a greater probability of delamination [20].

Several processing parameters influence splat morphologies - powder characteristics (chemistry, morphology, structure), plasma torch design (nozzle diameter, powder injection location, injection angle), arc current, arc voltage, spray distance, powder feed rate, carrier flow rate, and substrate preheat temperature [21–24].

To optimize the properties of the coatings, the plasma process parameters must be optimized. The entire process of the spray parameter development, microstructure evaluation, and the study of the properties of the coatings is determined experimentally and is iterative in nature. So, to better understand the effect of the processing parameters on the properties of the coating structure, computational methods are utilized.

1.3 Computational Methods and Their Limitations

With many variables in terms of processing parameters, equipment, and powder feedstock, it is very challenging to simulate coating structure as well as the microstructures. There have been numerous computational studies for the simulation of splats for the variation in plasma processing parameters. The formation of tungsten droplets on a wavy surface was studied by Liu et al. [25]. Bussman et al. [26] designed a 3D numerical model to simulate the splashing of the droplet in isothermal conditions. Pasandideh- Fard et al [27] studied the influence of substrate temperature on splat morphology of Nickel splats. Jiang et al. [20] investigated the influence of substrate temperature on the splat geometries of Molybdenum on different substrate materials.

Likewise, there are some computational techniques that are studied to develop the coating structure. A two-dimensional model based on the Monte Carlo technique was developed by Knoteck and Elsing [28]. Chen et al. [29] formulated a ballistic method to

study coating growth and pore formation. Wei et al. [30] studied the formation of coating buildup using the LAVA3D-P program.

The available computational models are either focused on the simulation of the splats or the simulation of the coatings structure. There lacks a comprehensive computational model which can simulate both splats and the coating structure and predict the properties of the coatings. So, in this study, a comprehensive technique is introduced that can predict the microstructure as well as mechanical properties of the plasma sprayed coatings.

1.4 Research Objective

One of the areas of interest in the field of plasma spray is the development of a computational technique that can tailor the mechanical and thermal properties of the coatings. In this study a novel computational desktop manufacturing approach is developed that can predict the microstructure as well as mechanical properties of the coatings with the variation in the plasma processing parameters. In this scheme, splat microstructure is simulated for different spraying conditions, three-dimensional coating structure is formed using simulated splat morphologies and the mechanical properties of the coatings are predicted. The results from the computational approach are validated with the results from experiments to determine the robustness of the model developed. The approach will allow for the development of the novel coatings in a shorter amount of time, resulting in the reduced cost and better estimation of coating properties.

Figure 1-3. Schematic Diagram of Computational Approach

Figure 1-3 summarizes the research objective which includes three specific aims:

1. Computational Fluid Dynamics for the simulation of splats
2. The arrangement of the simulated splats to form a two-dimensional coating structure and stacking of two-dimensional images together to create a three-dimensional coating structure using a stochastic approach
3. Microstructure-based finite element analysis for the prediction of the mechanical properties

This computational approach can be used as the cornerstone in the development of a universal technique to predict the mechanical properties of the coatings. It needs to be emphasized that more than one hundred parameters are involved in plasma spray in terms of processing parameters, equipment, and starting powder feedstock. So, the development of the computational model can also reduce the cost of the development of the coatings as this approach allows for the tailoring of the coating properties with a reduced number of experiments. This approach can be further extended to other materials as well as other thermal spray processes such as HVOF (High-Velocity Oxygen Fuel).

In developing this thesis, six chapters are presented. Chapter one presents the introduction and background of the topics related with the plasma spray, their applications, and the benefits of developing a computational approach to predict the microstructures as well as the properties of the plasma sprayed coatings. The following chapter is Chapter 2, in which the state of art is presented. Some previous investigations on the determination of microstructure and mechanical properties of the coating structure using the empirical techniques and computational techniques are discussed. Chapter 3 details the experimental

procedure, and the computational scheme. Chapter 4 provides the validation of the results obtained from the computational techniques with the experimental studies. Finally, Chapter 5 presents the major conclusions and Chapter 6 sets some suggestions for future studies to expand knowledge in the field of computational studies of plasma sprayed coatings.

CHAPTER 2. LITERATURE REVIEW

The relationship between the microstructure and the properties of the plasma sprayed coatings is complex due to a large number of processing parameters- plasma current, plasma gas composition and flow rate, standoff distance, plasma gun transverse speed and powder parameters [31–34]. In this section, a brief overview of the effect of the spraying parameters in the formation of the splats and the coating structure is presented, along with the state of the art of the experimental techniques for the selection of the optimal processing parameters and computational techniques that are developed to predict the microstructure and properties of the plasma sprayed coatings.

2.1 Processing Parameters in Plasma Spray

The effect of some of the key- processing parameters in the formation of the microstructure and the properties of the coatings structure are discussed below:

1. Feed rate

The feed rate should be maintained in the plasma spray process. A higher feed rate increases the number of the particles in the plasma plume resulting in the increase of the unmolten particles and porous coating structure. The unmolten particles may also not adhere properly to the substrate and bounce back, resulting in the decreased deposition for higher feed rate [35]. The feed rate also affects the thickness of the coating developed.

2. Stand-off Distance

Stand-off distance or the spray distance is the distance between the tip of the gun and the substrate surface. At the larger stand-off distance, the particles become partially solidified before impacting the substrate resulting in the formation of the splashed or the

fragmented splats. And, at the lower spray distance, the particles may not have enough time to melt and may erode the surface of the substrate [36].

3. Substrate Temperature

Fukumoto and Huang discovered from their studies that morphologies of splats were highly dependent on the substrate temperature. The Transition temperature (T_c) is defined as the substrate temperature in which half of the splats are disk type, and the transition temperature is dependent on the coating material than the substrate material [37,38]. Sampath et al. estimated the transition temperature to be close to 10% of the melting point of the material [18]. Though the exact explanation of this mechanism is unknown, it is believed that at the transition temperature, the adsorbates, and the condensates are removed from the surface [38].

Figure 2-1. Nickel splat morphologies on AISI 304 substrate. (a) Splash splat, $T_s = 300$ K. (b) Disk splat $T_s = 673$ K. (c) Fraction change of disk splat with substrate temperature [37]

4. Plasma Gas Flow Rate

Plasma gas flow rate affects the microstructure and the properties of the coatings as it influences the dwell time as well as the trajectories of radially injected particles. With low gas flow rate, fine particles bounce off the plasma jet and are not melted and a porous coating structure is obtained. And, for a high carrier gas flow rate, the plasma temperature and the velocity of the particles is increased significantly causing the particles to escape the hottest region of plasma resulting in the low flow rate [39].

5. Substrate roughness

Substrate roughness is another critical parameter that affects the splat morphology. With the increase in the substrate roughness, the friction for the liquid droplet increases which results in the decrease of the flattening degree, forming more fingered splats [40].

6. Thermal Contact Resistance:

Pasandideh-Fard et al. [27] found that the rate of solidification is much more sensitive to the values of the thermal contact resistance than the substrate temperature. The value of the thermal contact resistance is affected by the gases and the solid impurities trapped at the particle – substrate interfaces [41]. If the thermal contact resistance is too low, cooling is rapid, so the splat begins to solidify. The solid layer obstructs and destabilizes the flow of the liquid resulting in the formation of fingered splats. If the contact resistance is too high, the particle remains liquid and spreads into a thin film, that ruptures internally forming splashed splat morphologies [42].

To understand the complex thermo-physical phenomenon occurring during the plasma spray process, several studies have been carried out. These studies can be grouped

broadly into two different techniques: Experimental techniques and computational techniques.

2.2 Experimental Techniques

A common way to study the effect of plasma spray processing parameters on the coating properties is to use a statistical design of experiments (DOE) approach to empirically relate the parameters to the properties [40,43]. The empirical technique involves changing one process parameter at a time to observe its effect on the microstructure and the properties of the coating [44]. In this process, several experiments are carried out to identify the optimum parameters required for tailoring the properties of the coatings. The first step in the design of experiments is to determine the choice of the significant process parameters. The parameters are fixed at low (-1) and high (+1) inside the experimental domain. The four major techniques utilizing the design of experiments in plasma spray techniques are:

1. Hadamard or Plackett-Burman matrices [40,45]
2. Two-level full factorial design (2^k) [45,46]
3. Two levels fractional factorial design (2^{k-m}) [43]
4. Response of surface methodology (RSM) designs [47]

Many experimental studies are carried out to understand the effect of different spraying parameters on the microstructure and the properties of the plasma sprayed coatings. McPherson [48] studied the relationship between the microstructure and the mechanical properties of the coatings. The mechanical properties of the coatings were found to improve with the increase in contact between lamellae and between lamellae and substrate. Kulkarni et al. [15] studied the correlations between the processing and the

porosity and properties of yttria-stabilized zirconia coatings. With the increase in the size of the particles, fragmented and fingered splats were observed resulting in the increase of the porosity of the coatings leading to lower thermal conductivity and elastic modulus. Also, the increased substrate temperature resulted in the formation of disk-shaped splats resulting in the enhanced coating properties. Sampath et al. [18] studied the effect of the substrate temperature on the microstructure and properties of yttria-stabilized zirconia coatings. The study confirms that at the threshold temperature of around 10% ~ 250- 300°C of the melting temperature of zirconia, the splat morphologies change from fragmented (splashed) to disk-shaped. At the higher substrate temperature, the inter-splat and intra-splat contact are improved resulting in the reduced porosity and increased strength.

Figure 2-2. Morphologies and 3-D surface profiles of zirconia splats prepared on (a) and (c) cold substrate (100°C) and (b) and (d) hot substrate (250 – 300°C) [18]

Bianchi et al. [46] splat morphologies of zirconia at two different substrate temperatures. Disk-shaped splats with low thermal contact resistance were observed for the hot substrate and the fragmented splats were observed for the cold substrates. The adhesion-cohesion of the splats was found to be almost three times for the hot substrate over the cold substrate. Wang et al. [47] studied the effect of the splat interfaces in the effective properties of the plasma sprayed zirconia coatings. They observed that the splat interfaces result in 25 -60% of the total reduction in the effective properties of the coatings. The splat interfaces were observed to have greater influences in the thermal conductivity than on the elastic modulus. Jiang et al. [20] studied the splat morphologies of Molybdenum for different substrates at different temperatures. They found that splat morphologies changed from fragmented to the disk-shaped with the increase in the substrate temperature. The lamellar structure with fewer pores was observed for the increased substrate temperature. The thermal conductivity and hardness were found to increase at a higher substrate temperature.

Using the experimental techniques, determination of the relationship between the processing variables and the resulting particle characteristics and properties of coatings is very tedious, time-consuming and expensive. Also, the reproducibility of the coatings formed from the experimental routes is also one major challenge in the field of plasma spray [49].

2.3 Computational Techniques

Several computational techniques are used in the modeling of the particle flow in plasma spray processes such as Eulerian-Lagrangian model, and Eulerian-Eulerian approach [50]. In Eulerian approach, a control volume is defined and the flow of the

material through the control volume is analyzed. In Lagrangian approach, the properties are determined by tracking the motion of the particles. In the plasma flow process, for the Eulerian-Lagrangian approach, the Lagrangian model is used for the simulation of the particle phase, and the gas is modeled as a continuum phase [51]. In the Eulerian-Lagrangian model, the force controlling the motion of the particles is the drag. The Lagrangian models are classified as stochastic and deterministic models. The stochastic model considers the gas turbulence effects while the deterministic models neglect the gas turbulence effects [52]. In the Eulerian-Eulerian approach, the particles, as well as the gas, are modeled as a continuum [53]. Interface tracking methods such as Volume fraction method (VOF) are also used for the prediction of splat morphologies and coating build up in the plasma spray processes. Various computational tools that use either one or the combination of those above-mentioned techniques have been used by researchers to simulate splat formation and the coating structure to understand the effect of the processing parameters on the overall properties of the coatings.

A large amount of research has been conducted on the impact and solidification of the molten droplet on the substrate. Madejski developed a two-dimensional radial flow model to study the degree of flattening of molten droplet upon impact in 1976 [54]. A two-dimensional model that investigated the droplet impact velocity and temperature, the effect of the substrate and its temperature and the effect of the thermal contact resistance was developed by Bennett and Poulikakos [55]. Zhao et al. [31,56] used both experiments and a two-dimensional numerical model based on the Lagrangian approach to study the fluid dynamics and heat transfer during the impact of the liquid droplets. Pasandideh-Fard et al. [57] simulated the impact and solidification of tin droplets on a steel plate and validated

the results with experiments. Bussmann et al. [26] developed a three-dimensional computational model for the simulation of impacting droplet at the isothermal condition. Ghafouri -Azar et al. [58,59] studied the interactions between the splats using a three-dimensional model. The volume of Fluid (VOF) algorithm was used to track the free surface deformation.

The formation of the coating is very complex in nature. Konteck and Elsing [28] developed a two-dimensional computational model to calculate the temperature distribution in the coating and substrate. The thermal conduction equation is solved for the coating subsegments according to the growth law. The study predicted the size and distribution of interlamellar cracks and pores in the coating. Cirolini et al. [60] formulated a two-dimensional stochastic model and postulated the interactions of the splats deposited on top of others. The model shows how the complex features of the plasma sprayed coatings depend on the process of its deposition. Harding et al. [61] studied the formation of a two-dimensional computational model to build up the coating structure and correlate the properties of the coatings with the process parameters. Kanouff et al. [62] studied the computational modeling of coating inclined at an angle to the substrate and calculated the surface roughness of the coating. String method was used for the modeling of coatings in which the equally spaced node points define the shape of the coating structure and the change in the shape is tracked as the deposition is made. This stochastic model was capable of the deposition of a large number of thermal spray droplets and computing the surface roughness of the coatings.

Hansbo and Nylén [63] developed a spray deposition model to simulate the coating layer build up. An iterative method was used to simulate the robot motion. Chen et al. [29]

developed a two-dimensional ballistic deposition model to study coating growth and pore formation. The deposition of the particles using plasma spraying process was assumed to follow a Gaussian distribution and the computer program was developed to simulate the growth of the coating and generation of the pores based on the droplet deformation. Wei G. et al. [64] studied the formation of the coating buildup using the LAVA3D-P program. The 3D model developed was based on the probability density function. Ghaufouri et al. [65] developed a three-dimensional stochastic model capable of predicting the porosity, thickness, and roughness of the coatings as a function of process parameters. Several equations were formulated in this study to calculate the splat sizes after the droplet impact and the interactions of the splats with one another. The porosity is assumed to be due to the curling of the splats and the three-dimensional cartesian grid with the volume tracking VOF algorithm was used.

As discussed, different computational techniques and tools have been utilized for the simulation of the microstructure and the coatings for plasma spray. The complete modeling of the plasma spray involves three distinct processes: spray particles, impact and splat formation and the deposition of the coating. Despite the scientific and technical progress, there is a lack of understanding of the process-microstructure- property relationship. Several computational fluid dynamics analysis of the gas and the particle dynamics has been performed but the research works for the complete modeling of the plasma spray from the spray particles to the formation of the bulk coating structure is still in the rudimentary stage. Furthermore, the present computational studies are limited to the simulation of either only the microstructures or the coating structure. So, this work aims to take the modeling of the plasms spray process further by studying the formation of the

microstructure, as well as the coating structure and predict the mechanical properties of the coatings. In this study, a computational model approach is developed to predict the effect of various spray parameters on the microstructure as well as the properties of the bulk coating.

CHAPTER 3. MATERIALS AND METHODS

This chapter discusses in detail the experimental and computational methods used to synthesize and characterize the plasma sprayed aluminum oxide splats and coatings. The processing parameters, characterization techniques, and tools for the microstructural and mechanical evaluation of the coatings are discussed. The various computational tools used for the simulation of the microstructure and coating structure are presented.

3.1 Experimental Methods

3.1.1 Processing

3.1.1.1 Powder

Aluminum oxide is quite abundant and relatively inexpensive material making it ideal for the plasma spray applications due to its abrasion- resistance, chemical inertness, resistance to thermal shock and mechanical strength at high temperatures [66,67]. Aluminum oxide (Al_2O_3) powder from Praxair (AlO-101) with the size distribution of 15-45 μm was used in this study.

3.1.1.2 Substrate

Low carbon steel coupons of dimensions 50 mm $\times$ 19 mm $\times$ 3.2 mm was used as the substrate material. The roughness of the substrates was measured by an Optical Profilometer (Nanovea PS50, Irvine, California, USA). The substrates were polished to obtain roughness values (R_a) less than 0.1 μm for obtaining the splats. Heat gun (STEINEL, HG 2510 ESD, MN, USA) was used for preheating the substrate. The substrate temperature was continuously monitored with the thermocouple (KMQSS-020U, Omega Engineering Inc.) that was placed by drilling through the substrate thickness.

For the deposition of the coatings, the substrate was grit blasted ($R_a \sim 2\text{-}3\ \mu\text{m}$). The substrate temperature was monitored using the infrared pyrometer (IR) (Raytek Raynger MX®) and the deposition of the coatings was done when the substrate temperature reached the desired values.

3.1.1.3 Processing Parameters

The effect of the two processing parameters: power and substrate preheat temperature on the microstructure and the properties of the coatings were investigated in this study. Three different powers: 28 kW, 32 kW, and 35 kW and three different substrates preheat temperatures: 100°C, 180°C, and 250°C were used. The range of the power and the substrate preheat temperature were taken from the literature [66,67].

For the variation of the power, the current was changed keeping the voltage constant at 40 Volts. Plasma spraying of three powders (A-SD, A4C-SD, and A8C-SD) was carried out using Praxair SG 100 plasma gun (Praxair, Danbury, CT, USA) on low carbon steel substrate. Powders were carried by argon gas through Praxair model #1264 powder feeder and internally injected in the plasma gun. And the plasma gun movement was maintained at 25 mm/sec.

Table 3-1. Processing parameters used in the formation of splats and coatings

Primary	Secondary	Stand-off from the substrate	Powder feed
Gas, Argon	Gas, Helium	(mm)	rate
(slm)	(slm)		(g/min)
56.6	59.5	100	3.7

3.1.1.4 Formation of Splats and Coatings

For obtaining single splats, a shield plate with the holes of 2 mm diameter was placed in front of the plasma gun aligned in the direction of the spray to collect the single layer of splats without overpopulating the substrate. The substrate with the roughness less than $1\text{ }\mu\text{m}$ was placed behind the shield plate at the distance of 25 mm. The distance between the shield plate and the plasma gun was 75 mm maintaining the total spray distance of 100 mm. For obtaining coating structures, the same spray parameters used for the synthesis of the splats were used. About 8-10 passes on the grit blasted steel substrate was done to obtain about 150- 200 μm thick coatings. The schematic diagram for the experimental setup is shown in Figure 3-1.

Figure 3-1. Schematic Diagram for Plasma Spray Setup

3.1.1.5 In-flight Particle Diagnostic Sensor

For measuring the inflight particle temperature and velocities an in-flight diagnostic sensor AccuraSpray™ in-flight diagnostic sensor (Tecnar Automation Ltée, QC, Canada) was used. Accuraspray diagnostic system provides ensemble average data which represents the particle properties that are measured in the volume of approximately 75 mm³. The diagnostic sensor measures the time of the travel of particles over two closely placed optical fibers to determine the particle velocities. The electronic pulses generated by the photomultiplier to identify the particles and calculate the time of travel for a certain distance. These signals are then cross-correlated with the precision of 99.5% to measure the particle velocities. The temperature of the in-flight particles is measured by two-color pyrometry. The range of the particle velocities and temperatures that can be measured with the diagnostic sensor are 5 -1200 m/s and 1300 – 4000°C with the cross-correlation factor > 0.9. CCD camera in the diagnostic system enables the analysis of the plume appearance [68,69].

Figure 3-2. Operational Principle of AccuraSpray Diagnostic Sensor [68]

3.1.2 Characterization

3.1.2.1 Optical Microscopy

The morphology of splats was studied using an Optical Microscope (VERSAMET 3 Metallography Microscope). A total of 80 splats from 6-7 images were analyzed for each spray condition. The splat size distribution and the volume fraction of splat shapes (disk, fingered, and fragmented) were determined from the optical micrographs. For the fragmented and fingered splats, the diameter of the splats was measured from the center to the end of the finger. The volume fraction of the splats was defined as the ratio of the number of splats with specific morphology to the total number of the splats analyzed.

The coatings were cut, mounted and polished and the cross-sectional images of the coatings were captured with the optical microscope. About 3-5 images of the coatings for each spraying condition was obtained to study the porosity distribution.

3.1.2.2 Porosity Measurement

The porosity of the coatings was measured by a public domain image processing software ImageJ [70,71]. All the images of coatings were threshold appropriately and converted into binary images to calculate the porosity. The similar image analysis technique is reported in the computation of the porosity of the coatings [72]. Around 4-5 optical images of the coatings were used for the calculation of the porosity to compute the relative porosity of the coatings for each spraying condition.

3.1.2.3 Nanoindentation

Nano-indentation tests were conducted to obtain the elastic modulus from the polished cross-sectional area of the Al_2O_3 coatings using TI 900 Triboindenter (Hysitron Inc., Minneapolis, USA). A 100-nm diameter diamond Berkovich tip was first calibrated

with a standard fused silica sample, by fitting the contact area with a polynomial function of contact depth. Tests were carried out in a quasi-static load control mode. The peak load of 8500 μN was applied. The elastic modulus was obtained from the unloading curve from nanoindentation using Oliver-Pharr method [73]. At least 25 indents were performed on each sample.

3.2 Computational Methods

3.2.1 Simulation of Single Splats Using Computational Fluid Dynamics Approach

Computational fluid dynamics software, SIMULENT- DROP (SimDrop) developed by Simulent Inc., Toronto [74] was used for the simulation of splats. SimDrop employs a three-dimensional Eulerian fixed grid algorithm that utilizes volume tracking approach to track the deformation of fluid. The Navier- Stokes equation along with the heat transfer and phase change is solved using a finite difference scheme. VOF tracking algorithm is used to track the droplet-free surface.

The free surface of the deforming droplet is tracked by VOF scheme.

f is the fraction of the cell volume occupied by the liquid

$f = 1$, Cell is occupied by liquid

$f = 0$, empty cell

$0 < f < 1$, free surface

The equations of conservation of mass, momentum, and energy for the Newtonian, compressible and laminar flow are:

$$1. \text{ Conservation of mass: } \nabla \cdot V = 0 \quad (1)$$

$$2. \text{ Conservation of momentum: } \frac{dV}{dt} + \nabla \cdot (VV) = -\frac{1}{\rho} \nabla P + \nabla^2 V + \frac{1}{\rho} F \quad (2)$$

where V is the velocity vector, P is the pressure, ϑ is the kinematic viscosity, ρ is the density, F is the body forces (surface forces)

Neglecting the viscous dissipation and assuming that the densities of liquid and solid are constant at melting point and equal

3. Energy Equation:
$$\frac{\partial h}{\partial t} + (V \cdot \nabla)h = \frac{1}{\rho} \nabla \cdot (k \nabla T) \quad (3)$$

where h is enthalpy, k is the thermal conductivity, and T is the temperature

f is the fraction of the cell volume occupied by liquid, and the scalar function f is passively advected with the flow, f should satisfy the advection equation

4. Advection Equation:
$$\frac{\partial f}{\partial t} + (V \cdot \nabla)f = 0 \quad (4)$$

Young's algorithm is used to track the free surface. The algorithm consists of two steps: approximate reconstruction of the surface and the geometrical evaluation of volume fluxes across the cell faces. Surface tension effects are included in the body force acting on the fluid surface by using the Continuum Surface Force (CSF) method developed by Brackbill et al. [75].

For all the simulations, due to the presence of the symmetric boundaries, only a quarter of the domain was considered. This helped to save computational time due to the reduction of the problem size. Along the symmetric boundaries, free slip, no penetration conditions, and adiabatic thermal boundary conditions were applied. Similar use of computational fluid dynamics approach for the simulation of hydrophobicity of the water droplets in lotus leaf [76], impact of molten Nickel and Zirconia on different patterned surfaces of silicon [77], simulation of splat morphologies of the Aluminum oxide reinforced CNTs [78], and

simulation of splat morphologies of the spray dried Aluminum Oxide- graphene nanoplates [79] has been reported by the researchers.

The simulation of splats was performed for the spraying conditions mentioned in section 3.1.1.3. Seven different powder size distribution of 15, 20, 25, 30, 35, 40 and 45 μm were used for the simulation. From the previous studies on the crystallographic phases of the aluminum oxide powder and plasma sprayed coating structure, it was found that only a trace amount of gamma phase is present [80]. So, in this study, it is assumed that the crystallographic phase of the aluminum oxide remains the same (alpha-phase) throughout the entire plasma spray process.

The thermo-physical properties of aluminum oxide obtained from the literature were taken as the input parameter and listed in Table 3-2 [78].

Table 3-2. Thermo-physical properties of Aluminum Oxide (α -phase) for splat simulation [78]

Property	Value
Density(kg/m ³)	3990
Liquid phase kinematic viscosity (m ² /s)	1.026×10 ⁻⁵ at 2437 K
Liquid phase thermal conductivity (W/mK)	7.86 at 2327 K
Liquid phase specific heat (J/kgK)	1358 at 2327 K
Liquid phase surface tension (kg/s ²)	0.69 at 2327 K
Solid phase thermal conductivity (W/mK)	36.16 at 298 K 5.9 at 1312 K
Solid phase specific heat (J/kgK)	772 at 298 K 1273 at 1312 K 1358 at 2327 K
Melting point (K)	2327
Heat of fusion (J/kg)	1.16×10 ⁶

Thermal contact resistance at the droplet-substrate interface was assumed as 10⁻⁷ m²K/W for 180 and 250 °C and 10⁻⁶ m²K/W for 100 °C [78,79,81,82]. The average particle temperature and velocity measured by the in-flight diagnostic sensor were taken as input parameters for the simulation of splats. The particle velocities and temperature for different power are presented in Table 3-3.

Table 3-3. Particle velocities and temperatures for different plasma power obtained from the in-flight diagnostic sensor during splat formation

Power (kW)	Particle velocity (m/s)	Particle Temperature (° C)
28	270	2477
32	277	2492
35	287	2497

The particle velocities, as well as the temperature, were found to be increased with the plasma power. To account for the cross-correlation factor of the measurement accuracy from the in-flight diagnostic sensor, the particle velocities and temperatures were varied by $\pm 10\%$ during the simulation of splats.

The resolution of the simulation in SimDrop is defined in terms of cell per radius (CPR). CPR is defined as:

$$\text{CPR} = \frac{\text{Diameter of the impinging droplet}}{\text{Calculation domain}} * \text{Number of grids (6)}$$

The higher the value of the CPR, the higher is the resolution of the grid. But, increasing the CPR value increases the number of grids resulting in the requirement of a large amount of computing time. The CPR value of at least 10 is recommended to obtain the solutions [73]. For choosing the optimum value of CPR for simulation, mesh independent study was carried out. Spread ratio, i.e., the ratio of the final splat size to the initial powder size (30 μm) was plotted for different CPR values as shown in Figure 3-3. It was observed that for the CPR value of 15, the change in the spread ratio is less than 5%.

So, the CPR value of 15 was used in the SimDrop software for the high-resolution simulation of one-quarter of splat to reduce the computational time.

Figure 3-3. Maximum spread ratio for different values of CPR

3.2.2 Formation of Three-dimensional Coating Structure Using Stochastic Models

After obtaining the splat morphology computationally, the next step was to create a three-dimensional coating structure by stacking simulated splats. The splats were first arranged in a two-dimensional grid structure to obtain a two-dimensional image. It was assumed that the spreading of a splat depends on the previously deposited splat adjacent to it. The images from SimDrop were arranged according to the splat geometry and distance between the splats based on the stochastic model developed by Ghafouri-Azar et al.[65]. Ghafouri-Azar had developed four different possible conditions to study the shape of the

second splat formed due to the interactions between two splats; previously deposited splat and impacting splat. The distance between the neighboring splats that determine the shape of the splats as per the stochastic model developed by Ghaufori-Azar et al. [65] is presented in Table 3-4. 'R' is the radius, and 'Dn' is the diameter of the impacting splat under consideration. Dm is the diameter of the splat nearest previously deposited splat. 'l' is the smallest distance between the droplet impact point and the center point of the previously deposited splats. The shapes were determined based on the equations from the stochastic model. The ratios of the parameters a and b were obtained from the measurements and are presented in Appendix Table 1 - Appendix Table 9.

For the creation of the two-dimensional image, the volume fraction of splat morphologies (disk-shaped, fingered, and fragmented) and splat diameter distribution obtained experimentally were taken as the input parameters. Splats were arranged in a grid based on the distances as described in Table 3. A grid of 100 μm x 100 μm was created, and the simulated splats from SimDrop were arranged using a random walk probabilistic approach. Grid sizes were limited to 100 μm to minimize computational time and resources. The simulated splats were arranged randomly over the grid. The distances between the shapes were provided as the input parameter based on the shapes of the impacting splat morphology and previously deposited splat according to the four possible scenarios for the droplet interactions as referred in the stochastic model and Table 3-4 [65].

Table 3-4. Smallest distance (l) between two neighboring splats based on the equations describing splat shapes (Based on the stochastic model developed by Ghafouri-Azar et al. [65])

Equation No.	l (distance between two splats)	Equation Describing splat shapes	a/b
A	$0 \leq l \leq \frac{3}{8}Dm$	$\frac{x^2}{a^2} + \frac{y^2}{b^2} = 1$	$\frac{a}{b}$ $\approx 0.836 \left(\frac{l}{Dm} \right)$ $+ 0.6878$
B	$\frac{3}{8}Dm \leq l \leq \frac{1}{2}Dm$	$r^4 + a^4 - 2a^2r^2\cos 2\theta = b^4$	$\frac{a}{b}$ $\approx -1.2842 \left(\frac{l}{Dm} \right)$ $+ 1.2338$
C	$\frac{1}{2}Dm \leq l \leq \frac{7}{8}Dm$	$y = \frac{R}{e^{\frac{2x}{R}} + e^{-\frac{2x}{R}}}, (y > 0)$ $\frac{x^2}{a^2} + \frac{y^2}{b^2} = 1, (y < 0)$	1.0
D	$\frac{7}{8}Dm \leq l \leq \frac{Dm + Dn}{2}$	$\frac{x^2}{a^2} + \frac{y^2}{b^2} = 1$	1.1

To obtain the variation in the size of the splats, powder size of 15, 20, 25, 30, 35, 40, and 45 μm were simulated. And, the values of the velocity and temperature were varied as $\pm 10\%$ of the value measured from the in-flight diagnostic sensor in the experiments. As

presented in the stochastic model developed by Ghaufouri-Azar et al. [65] and Table 3, equations A and D were used for the arrangement of the disk and fingered splats, and equations B and C were used for the arrangement of the fragmented splat morphologies in a two-dimensional image. The input parameters taken for the formation of the 2D images are presented in the Appendix Tables 1-9. The 2D images were taken as an input in the MATLAB code where the images were stacked to form a three-dimensional coating structure. The various effects like “curling of the splats,” relieving of the residual stresses, micro-cracks, splat interfaces were not considered. The pores were assumed to be formed only due to the incomplete filling of the interstices.

3.2.3 Object-Oriented Finite Element Analysis for Elastic Modulus Prediction

The open source program Object oriented finite element analysis software, OOF2 developed by the National Institute of Standards and Technology (NIST), has the capability of predicting the macroscopic mechanical properties of the materials using microstructural features as the input [83]. The simulated coating’s cross-sectional microstructure was used as an input image for OOF2. The pixels from the image were first converted into two different groups based on the colors and two different groups are obtained- for materials and pores. A coarse mesh with a grid size of 50 x 50 pixels was made. The elements near the interfaces were refined and adjusted so that the interfaces were well described with element boundaries. For the mesh refinement, first, the edges are subdivided and marked and subsequently marked elements are replaced with smaller elements. This was done using the adaptive mesh refinement techniques, to refine the mesh. The mesh is generated by minimizing the energy function which is composed of the homogeneity part of the mesh and shape part of the element as described by equation 5.

$$E = \alpha E_{hom} + (1 - \alpha) E_{shape} \quad (5)$$

where α is a tunable parameter and varies between 0 and 1.

The homogeneity is the measure of how close an element is to enclosing a region that contains a single material. The shape energy E_{shape} (shape part of the element) is a measure of a feature of the shape of the element [84]. The quality of the mesh is determined by the homogeneity. The homogeneity of the images was obtained to be more than 98% for the mesh refinement, to ensure the grid independent and accurate solutions. During the mesh refinement, bigger elements are converted into smaller elements by moving the element nodes at the pixel boundaries using the snap tool. Smooth and anneal tools are utilized to move the nodes according to the selection criterion. The actual boundary of the mesh was obtained by the intense refinement of pixels at the boundary. OOF2 creates a finite element-mesh which reflects the shape of the different phases in the microstructure with the associated material parameters.

The values of elastic modulus for Aluminum oxide as 385 GPa and the Poisson's ratio as 0.23 were provided as the input parameter in the finite element analysis software [85]. The pores were considered as the viscoelastic material. The input values for pores were provided as 10^{-8} GPa for Elastic modulus and 0.48 for the Poisson's ratio [48].

After the definition of the material properties and the construction of the mesh, the boundary condition of a uniaxial tensile strain of 1% was applied in the positive Y-axis direction. 1 % strain value was chosen to be within the elastic regime of the material. After the strain was applied, the integral stress value at the boundary was measured for the uniaxial strain of 1%. And the elastic modulus of the coatings for different spraying conditions was computed.

CHAPTER 4. RESULTS AND DISCUSSION

This chapter examines the results obtained from both the experimental and computational methods. The results from the computational analysis are validated by the results of the experimental techniques to determine the robustness of the proposed computational approach.

4.1 Formation and Simulation of Splats

In this study, the effect of power and substrate preheat temperature on the formation of plasma sprayed Aluminum splats were studied using both experiments and simulation. Simulated splat morphologies are obtained from the SimDrop software. The splat morphologies obtained from simulation and experiments are compared in Figure 4-1 to 4-3.

Figure 4-1. Comparison of splat morphology obtained from experiments and simulations, a) Power 28 kW, Substrate Preheat Temperature 100 °C, b) Power 28 kW, Substrate Preheat Temperature 180 °C and c) Power 28 kW, Substrate Preheat Temperature 250 °C

Figure 4-2. Comparison of splat morphology obtained from experiments and simulations, a) Power 32 kW, Substrate Preheat Temperature 100 °C, b) Power 32 kW, Substrate Preheat Temperature 180 °C and c) Power 32 kW, Substrate Preheat Temperature 250 °C

Figure 4-3. Comparison of splat morphology obtained from experiments and simulations, a) Power 32 kW, Substrate Preheat Temperature 100 °C, b) Power 32 kW, Substrate Preheat Temperature 180 °C and c) Power 32 kW, Substrate Preheat Temperature 250 °C

A larger number of fragmented and fingered splats were observed for lower power and substrate preheat temperature. This can be related to the localized solidification of the molten droplets which obstructs the spreading resulting in the formation of fingers and fragments. The results from the SimDrop software accurately captured all the intricate features like fingers and fragments of the splats. The splat morphologies from the simulations showed a great resemblance to the morphologies obtained from experiments.

The volume fraction of splat shapes (disk, fingered, fragmented) for varying plasma power and the substrate temperature was determined experimentally as shown in Figure 4-4. A total of 80 splats from 6-7 images were analyzed for each spray condition. The volume fraction of the splat shapes (disk, fingered and fragmented) for the variation in the processing parameters were determined experimentally. The volume fraction of the splat morphology was measured as the ratio of the number of the splats with specific morphology to the total number of splats analyzed.

Figure 4-4. Volume fraction of splats for a) 28 kW, b) 32 kW and c) 35 kW plasma power

Figure 4-4 shows that the volume fraction of the disk-shaped splats increased with the increase in power and substrate temperature. Disk-shaped splats increased four-fold from a mere 15% at 28kW, 100°C to 63% at 35 kW, 250°C.

A wide range of powder size 15-45 μm was used for the splat formation. So, to understand the size distribution of the splat size for the various spraying conditions, size distribution plot was made.

Figure 4-5. Splat size distribution for different substrate preheat temperatures for a) 28 kW, b) 32 kW and c) 35 kW plasma power

Figure 4-6. Average diameter of different splat morphologies for a) 28 kW, b) 32 kW and c) 35 kW plasma power

From the figures 4-5 and 4-6, it is observed that the diameter of the splats, as well as the number of the splats with larger diameter increases with the increase in power as well as substrate, preheat temperature.

The power directly affects the degree of the melting of the powder particles. Higher power results in the better spreading of the molten droplets, thus increasing the population of the splats with higher splat diameter. So, with the increase in the power and substrate temperature, the splats with the larger diameter for all splat morphologies- the disk, splashed and fragmented were observed.

Splat size, as well as the volume fraction of the disk-shaped splats, increased with an increasing plasma power and substrate temperature. This can be explained by the improved wetting (or lower viscosity) that occurs between the molten droplet and the substrate allowing for a better spread at higher substrate temperature [86]. With the increase in plasma power, both the in-flight particle temperature and velocity increases allowing the particle to spread uniformly increasing disk-shaped splats. Increase in the substrate temperature delays the localized solidification allowing the molten droplet to spread uniformly resulting in the disk-shaped splats.

Although the exact mechanism and the effects of the substrate preheat temperature on the splat morphologies are unknown, it is believed that the enhanced wetting is also resulted due to the removal of the adsorbed gas and condensates [38].

At the higher substrate temperature, better contact and uniform heat and heat conduction can lower the chance of local solidification or delay the initiation of the solidification. According to Vardelle et al.[87], preheating the substrate above the transition temperature ensures that the temperature of the molten liquid droplet remains

above the hyper-cooling temperature and the splat remains in the liquid phase resulting in the uniform solidification.

4.2 Simulated and Experimentally Obtained Coating Microstructure

The formation of a two-dimensional image and the stacking of the images to form a three-dimensional coating structure is shown in Figure 4-7.

Figure 4-7. Formation of three-dimensional coating structure from simulated splats

From the literature, it is found that the first layer of the splats deposited on the substrate play a vital role in the coating build-up process. Jiang et al. [88] suggested that the spreading of the splats deposited on top of the previously deposited splats follow the similar pattern as that of the spreading of the droplet on the substrate or previously deposited splat layer during the coating build-up process. In, this study, the first layer of the splats deposited on the substrate is stacked to form a three-dimensional coating structure with a thickness of 100 μm . The thickness was chosen such that the obtained computational coating is thick enough to be considered a bulk coating for computing porosity and mechanical properties.

The comparison of the cross-sectional images of the coating structure from experiments and computational routes is shown in the figures 4-8.

Figure 4-8. Cross-sectional image of coating, a) from experiments power 32 kW, substrate preheat temperature 180 °C and b) from computational stacking power 32 kW, substrate preheat temperature 180 °C.

The porosity of the coatings obtained from experiment Figure 4-5 (a) was measured using image analysis software ImageJ and the porosity of the simulated coatings Figure 4-6 (b) was measured using finite element analysis (FEA). The porosity from the finite element analysis are assumed to be formed only due to the incomplete filling of the splats and are more elongated or plate-shaped in nature, while the porosity of the experimentally obtained coatings is more globular or round due to the curling of the splats. Figure 4-9 helps to understand better how the porosity differs for the real-time plasma spray deposition process and the computational approach.

Figure 4-9. Deposition of real-time plasma process and computational model

In the real-time plasma process, the splats curl during the solidification process. Not, only this the entrapment of the air or the gas bubbles, release of the thermal stress during the cooling, imperfect contact, and stress relaxation causing the intra-splat cracks results in the formation of the globular or spherical pores. For, the computational model, a unit thickness is assumed for the 2D images during the stacking process in MATLAB code resulting in the plate-shaped or elongated thickness.

The porosity values obtained from the experimental and simulated routes are compared in Figure 4-10.

Figure 4-10. Comparison of the porosity of the cross-sectional orientation of the coatings obtained from finite element analysis (FEA) for the computational coating structure and ImageJ for the experimental coatings.

The average values of the porosity in the coatings obtained from experiments were measured from 4-5 different images for each spraying condition. It is observed that experimental porosity values are within 1-2% error margin of the porosity in the simulated coatings. The marginal difference in the porosity values can be attributed to the

assumptions that were made during the stacking of the image. Intersplat porosity and curling of the splats were not accounted for in the simulated coating.

Figure 4-10 shows that the porosity of the coating reduced from 15.5 % for 28 kW, 100°C ~ 8.5 % for 35 kW, 250°C. The increase in power and substrate preheat temperature resulted in the reduced porosity for the coatings obtained both by the computational and experimental routes. This can be related to the increase in the fraction of disk-shaped splats which improves the packing. This also substantiates the findings of the increased volume fraction of disk-shaped splat morphologies for the increase in power and substrate preheat temperature. With the increased in the number of the disk-shaped splats the inter-splats adhesion and cohesion is enhanced resulting in the formation of dense coating structure.

4.3 Computed and Experimental Elastic Modulus of the Coating

Object-Oriented Finite Element Analysis software (OOF2) was utilized to compute the elastic modulus of the coating and compared with experimental values obtained using nanoindentation.. The various steps involved in obtaining elastic modulus from object oriented finite element analysis are shown in Figure 4-11.

Figure 4-11. Finite element analysis, acquiring an image, adaptive meshing, and application of boundary conditions.

The results from the nanoindentation and finite element analysis are compared in Table 4-1 below.

Table 4-1. Elastic modulus obtained from nanoindentation and OOF2 for the cross-section

Substrate	Power					
	28 kW		32 kW		35 kW	
preheat						
temperature	Nano	Finite	Nano	Finite	Nano	Finite
	indentation	Element	indentation	Element	indentation	Element
	(GPa)	Analysis	(GPa)	Analysis	(GPa)	Analysis
		(GPa)		(GPa)		(GPa)
100 °C	255 ± 14.3	297.7	263 ± 17.6	307.8	270 ± 15.1	308.2
180 °C	268 ± 34.9	303.6	271 ± 22.5	312.9	274 ± 13.9	316.6
250 °C	272 ± 16.8	307.1	280 ± 19.7	320.2	285 ± 18.4	328.4

The values of the elastic modulus in cross-section of the coating predicted by the microstructure based OOF approach are ~12% greater than experimental values.

This discrepancy between the predicted and experimental values can largely be attributed to the inability of the computational tools to perfectly capture inter-splat effects such as (i) inter-lamellar porosity, (ii) inter-splat microcracks, and (iii) curling of the splats.

Since these features are fine and thin, inter-splat cracks could not be modeled effectively in the finite element analysis resulting in an overestimation of the elastic modulus. Moreover, the occurrence of splat-sliding in the thermally sprayed coatings during nanoindentation results in the lower elastic modulus [89,90]. Splats slide past each

other resulting in the decrease of the resistance to deformation. To account for these differences caused by splat related effects, we introduce “inter-splat correction factor,” f_{is} . This factor establishes a quantitative comparison of elastic modulus obtained from computational modeling vis-à-vis experimental values as shown in equation 7.

$$E_{expt} = f_{is} * E_{comp} \quad (7)$$

Where E_{expt} is the elastic modulus determined by nanoindentation, E_{comp} is the elastic modulus of the cross-section determined by the object-oriented finite element analysis, and f_{is} is the inter-splat correction factor.

To compute the inter-splat correction factor, the line of best fit was determined using the ordinary least squares method regressing the values of elastic modulus from the finite element analysis as the variable (x) and the values from the nanoindentation as the variable (y). The method of linear calibration regression employed by Rosner et al [91] was used to compute the inter-splat correction factor (f_{is}) as 0.89.

The values of the elastic modulus obtained from the finite element analysis were corrected using the correction factor and compared with the values of the elastic modulus obtained from the nanoindentation as shown in the Table 4-2.

Table 4-2. Elastic modulus obtained from nanoindentation and OOF2 with an inter-splat correction factor for the cross-section

Substrate preheat temperature	Power					
	28 kW		32 kW		35 kW	
	Nano indentation (GPa)	OOF Analysis (GPa)	Nano indentation (GPa)	OOF Analysis (GPa)	Nano indentation (GPa)	OOF Analysis (GPa)
100°C	255 ± 14.3	266	263 ± 17.6	271	270 ± 15.1	274
180°C	268 ± 34.9	271	271 ± 22.5	278	274 ± 13.9	282
250°C	272 ± 16.8	274	280 ± 19.7	285	285 ± 18.4	292

The results from the computational approach and experiments are comparable within 3-5% deviation which depicts the ability of the computational approach to accurately predict the mechanical properties. Not, only this, the approach can also be used for the prediction of the thermal properties of the coatings [83].

CHAPTER 5. CONCLUSIONS

In this thesis, a novel multi-step computational approach is developed for predicting the effect of spray parameters on both the microstructure as well as the mechanical properties of the plasma sprayed coating structure. The volume fraction of the disk-shaped splats, as well as the porosity and the elastic modulus of the coatings, is improved with the increase in the plasma power and substrate preheat temperature.

The computational models are largely successful in predicting the mechanical properties of the ceramic coatings with some deviation ($\sim 12\%$) from experimental results. The deviation between experimental and computational results is attributed to the limitation of the computational models which does not account for intersplat features as microcracks, curling of splats, and splat-sliding. An inter-splat correction factor (f_{is}) was introduced to overcome the limitations of the computational model. After the correction factor, the elastic modulus values from computations are observed to be in reasonable agreement with the experimental values.

This study demonstrates that the proposed computational scheme can predict the mechanical properties of the coatings with a reduced number of experiments for various spraying parameters. This desktop design approach can be further extended to other materials for the development of coating structure with predictable properties and would be of immense importance to the thermal spray community.

CHAPTER 6. RECOMMENDATIONS FOR FUTURE WORK

In this work, a computational model has been successfully developed to predict the microstructure as well as the mechanical properties of the plasma sprayed coatings. Based on the conclusions of this research work, several recommendations for future work are made:

Limitations	Recommendations
1. Simulation of the splat morphology <ul style="list-style-type: none">• The properties of the particle such as surface tension, viscosity, thermal conductivity is taken as the input parameters which are not well known for various materials at high temperature range• Constant thermal contact resistance is assumed during the simulation of splats• Formation of oxide during the splat formation has not been considered in this model• The particle velocities and temperature used for the simulation of splats in this study	<ul style="list-style-type: none">• Inclusion of predictive analytical model to compute thermal contact resistance for variation in time and position would make the model more realistic• Addition of the study about the formation of the oxide, thickness of the oxide layers, and its subsequent effects in the morphology of the splats should be investigated• Study of the composition of the plasma gas including the temperature and velocity distribution, and plasma-particle interactions would allow for the

<p>was obtained from the in-flight diagnostic sensor, which provides a single data point for the measurement of particle velocity and temperature and the values were varied $\pm 10\%$ to account for the cross-correlation factor of sensor</p>	<p>prediction of the temperature and velocity of powder particles without the use of in-flight diagnostic sensor to make the model autonomous and robust</p> <ul style="list-style-type: none"> • Use of the computational fluid dynamics software to obtain computed shape data by varying particle temperature & velocities and comparing with experimental data for the validation
<p>2. Simulation of three-dimensional coating structure</p> <ul style="list-style-type: none"> • Size distribution and volume fraction of splats from experiments are taken as the input parameters for the formation of 2D image which are stacked together to form a simulated coating structure • The 2D image is obtained using simulated splat morphologies 	<ul style="list-style-type: none"> • Comparison of the size distribution and volume fraction of splats from experiments and the computational model (simulation of large number of splats) would provide an insight on the porosity as well as mechanical properties of the coatings • Inclusion of the roughness value for the grit blasted surfaces from the literature for the splat

<p>deposited on the polished substrate</p> <ul style="list-style-type: none"> • During the coating buildup process, the 2D images are stacked on top of each other for the formation of three-dimensional coating structure • A single layer of splats deposited on the substrate are stacked to form the coating structure in the model • The buildup of the coating also doesn't account for the splat related effects such as interlamellar pores, intra-splat cracks and splat boundaries • The phase transformation has not been accounted in this study 	<p>simulation for the coating buildup would make the model more precise</p> <ul style="list-style-type: none"> • The inclusion of effects such as “curling of splats”, difference in the thickness of the splats during the coating build-up should be explored which would enhance the model's predictive ability • The study of the deposition of the first layer of splats on the substrate and the subsequent deposition of the next layers on the material itself would augment the model • The effects of diffusion and phase transformation during the formation of coating structure should be explored with the help of thermochemical tools
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<p>3. Prediction of mechanical properties using oriented finite element analysis approach</p> <ul style="list-style-type: none"> • The finite element-based approach is unable to account for the boundary or the interface related effects • The input taken for the prediction of the mechanical properties are the 2D images which may not accurately represent the orientation of the pores in the coating structure 	<ul style="list-style-type: none"> • Use of the alternative approaches to study the three-dimensional coating structure for the prediction of the mechanical properties with the consideration of the micro and Nano-pores would make the model more reliable • Study of a different material would provide an insight into the robustness of the technique developed
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APPENDIX

APPENDIX Table 1. Parameters used for formation of 2D image for Power 28 kW and substrate temperature 100 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
15-20	2	1) 18.7	1) 0.97	3	1) 19.7	1) 1.02	4	1) 15.3	1) 1.02
		2) 17.6	2) 1.03		2) 18.6	2) 1.21		2) 18.6	2) 2.30
					3) 16.4	3) 1.14		3) 19.3	3) 1.70
								4) 17.7	4) 1.23
20-25	3	1) 22.7	1) 1.11	4	1) 23.4	1) 0.93	3	1) 24.7	1) 1.20
		2) 23.4	2) 0.82		2) 24.8	2) 0.89		2) 23.1	2) 1.07
		3) 21.7	3) 0.89		3) 21.6	3) 0.95		3) 20.4	3) 1.16
					4) 20.8	4) 1.13			
25-30	1	1) 28.9	1) 1.02	2	1) 25.6	1) 0.87	3	1) 29.2	1) 1.30
					2) 27.8	2) 0.72		2) 26.7	2) 1.13
								3) 25.2	3) 1.15
30-35	2	1) 33.6	1) 1.14	5	1) 31.6	1) 1.04	6	1) 30.8	1) 0.90
		2) 34.7	2) 0.97		2) 30.2	2) 1.13		2) 31.2	2) 1.36
					3) 34.6	3) 1.12		3) 32.7	3) 1.28
					4) 33.6	4) 1.09		4) 33.9	4) 1.35
					5) 31.8	5) 0.88		5) 34.7	5) 0.87
								6) 34.3	6) 0.86
35-40	0	-	-	7	1) 36.2	1) 1.03	3	1) 38.9	1) 0.88
					2) 35.2	2) 1.19		2) 39.4	2) 0.92
					3) 38.3	3) 1.24		3) 37.1	3) 0.79
					4) 37.7	4) 1.12			

					5) 38.9	5) 0.98			
					6) 39.2	6) 0.87			
					7) 36.8	7) 0.91			
40-45	3	1) 42.3 2) 44.7	1) 1.21 2) 1.02	4	1) 44.7 2) 43.5 3) 41.7	1) 1.02 2) 0.84 3) 0.99	5	1) 43.4 2) 44.9 3) 41.3 4) 42.6 5) 42.9	1) 0.92 2) 0.86 3) 1.13 4) 0.84 5) 1.24
45-50	2	1) 46.5 2) 48.1	1) 0.87 2) 1.12	5	1) 48.9 2) 49.4 3) 45.2 4) 46.3 5) 47.4	1) 1.02 2) 1.13 3) 1.20 4) 1.17 5) 1.31	5	1) 48.4 2) 46.3 3) 45.9 4) 49.3 5) 45.4	1) 1.20 2) 1.13 3) 0.86 4) 0.97 5) 1.17
50-55	1	1) 54.6	1) 1.14	1	1) 50.2	1) 0.98	4	1) 51.8 2) 53.5 3) 54.9 4) 50.7	1) 1.16 2) 0.76 3) 0.85 4) 0.97
55-60	0	-	-	1	1) 55.7	1) 0.96	1	1) 58.2	1) 1.13

APPENDIX Table 2. Parameters used for formation of 2D image for Power 28 kW and substrate temperature 180 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
15-20	1	1) 19.4	1) 0.98	4	1) 15.4	1) 0.98	3	1) 16.6	1) 1.13
					2) 16.7	2) 1.13		2) 15.2	2) 1.03
					3) 18.3	3) 1.11		3) 18.4	3) 0.86
					4) 19.5	4) 0.86			
20-25	4	1) 20.3	1) 0.94	5	1) 20.2	1) 0.91	6	1) 22.3	1) 1.12
		2) 21.4	2) 0.89		2) 21.4	2) 0.87		2) 20.1	2) 1.18
		3) 24.8	3) 1.14		3) 22.3	3) 1.12		3) 24.5	3) 0.81
		4) 24.6	4) 1.17		4) 22.7	4) 0.91		4) 23.7	4) 0.78
					5) 24.6	5) 1.03		5) 22.8	5) 0.84
								6) 21.3	6) 1.04
25-30	1	1) 28.9	1) 1.02	4	1) 25.1	1) 0.87	2	1) 29.6	1) 1.14
					2) 26.4	2) 0.98		2) 28.4	2) 0.98
					3) 27.3	3) 0.89			
					4) 29.1	4) 1.12			
30-35	1	1) 30.5	1) 1.16	2	1) 33.5	1) 1.06	1	1) 30.8	1) 1.18
					2) 34.9	2) 0.98			
35-40	1	1) 38.4	1) 1.04	1	1) 39.4	1) 1.27	2	1) 37.6	1) 0.99
								2) 36.8	2) 0.81
40-45	8	1) 44.2	1) 1.02	5	1) 44.8	1) 0.95	1	1) 43.5	1) 0.76
		2) 40.6	2) 1.12		2) 43.7	2) 0.79			
		3) 43.2	3) 0.98		3) 44.9	3) 0.84			
		4) 44.1	4) 0.99		4) 40.2	4) 0.79			
		5) 44.7	5) 1.05		5) 41.3	5) 1.12			
		6) 40.8	6) 1.18						

		7) 41.8	7) 1.22						
		8) 41.1	8) 1.34						
45-50	7	1) 47.8	1) 1.19	8	1) 45.6	1) 0.90	1	1) 47.8	1) 0.98
		2) 46.3	2) 1.2		2) 46.7	2) 1.19			
		3) 45.1	3) 0.85		3) 47.4	3) 1.23			
		4) 48.8	4) 0.98		4) 48.9	4) 0.96			
		5) 49.5	5) 1.14		5) 49.1	5) 0.99			
		6) 48.2	6) 0.99		6) 49.8	6) 1.07			
		7) 47.3	7) 0.91		7) 46.3	7) 1.12			
					8) 48.3	8) 1.15			
50-55	3	1) 50.3	1) 1.02	1	1) 53.4	1) 0.93	1	1) 54.8	1) 0.89
		2) 53.4	2) 1.11						
		3) 57.8	3) 0.99						
55-60	1	1) 58.5	1) 1.07	1	1) 59.2	1) 0.94	2	1) 59.8	1) 0.81
								2) 56.3	2) 1.01
60-65	4	1) 60.2	1) 1.05	0	-	-	0	-	-
		2) 61.4	2) 1.13						
		3) 63.8	3) 1.21						
		4) 64.2	4) 0.87						

APPENDIX Table 3. Parameters used for formation of 2D image for Power 28 kW and substrate temperature 250 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
15-20	1	1) 18.4	1) 1.13	0	-	-	0	-	-
20-25	1	1) 21.6	1) 0.98	1	1) 22.3	1) 0.89	0	-	-
25-30	3	1) 25.6	1) 0.84	5	1) 25.3	1) 1.13	4	1) 25.2	1) 0.91
		2) 27.8	2) 1.03		2) 26.7	2) 1.05		2) 26.7	2) 0.89
		3) 29.2	3) 1.23		3) 27.8	3) 1.24		3) 27.4	3) 0.86
					4) 28.4	4) 1.15		4) 28.3	4) 1.13
					5) 29.6	5) 0.87		5) 29.4	5) 1.08
30-35	5	1) 30.2	1) 1.13	3	1) 34.6	1) 0.84	3	1) 34.2	1) 1.01
		2) 34.6	2) 1.08		2) 33.8	2) 1.21		2) 33.8	2) 0.93
		3) 33.7	3) 1.02		3) 31.4	3) 1.02		3) 30.7	3) 0.86
		4) 31.6	4) 0.99						
		5) 34.1	5) 0.87						
35-40	3	1) 35.4	1) 0.75	4	1) 37.2	1) 1.02	2	1) 39.7	1) 0.95
		2) 36.8	2) 1.12		2) 38.4	2) 0.97		2) 38.7	2) 1.01
		3) 38.4	3) 0.89		3) 39.5	3) 0.94			
					4) 38.6	4) 0.92			
40-45	7	1) 40.2	1) 0.87	4	1) 44.7	1) 1.01	1	1) 43.2	1) 1.03
		2) 41.3	2) 0.82		2) 43.2	2) 0.94			
		3) 42.8	3) 1.07		3) 40.3	3) 0.98			
		4) 44.8	4) 1.18		4) 42.8	4) 1.12			
		5) 42.1	5) 1.13						
		6) 40.8	6) 0.85						
		7) 41.3	7) 1.04						
45-50	7	1) 45.3	1) 1.01	6	1) 45.1	1) 0.98	5	1) 45.4	1) 1.09
		2) 46.7	2) 1.12		2) 46.4	2) 1.13		2) 47.8	2) 0.97

		3) 47.8	3) 1.13		3) 47.8	3) 1.20		3) 48.4	3) 1.03
		4) 44.9	4) 1.08		4) 48.6	4) 1.18		4) 49.9	4) 1.08
		5) 43.4	5) 0.98		5) 49.4	5) 1.17		5) 47.3	5) 1.2
		6) 45.8	6) 0.99		6) 45.3	6) 1.01			
		7) 46.1	7) 1.14						
50-55	4	1) 52.3	1) 1.12	0	-	-	0	-	-
		2) 53.6	2) 1.13						
		3) 54.8	3) 1.06						
		4) 50.5	4) 1.08						
55-60	3	1) 56.7	1) 1.14	2	1) 57.8	1) 1.21	0	-	-
		2) 57.8	2) 1.04		2) 58.9	2) 1.09			
		3) 59.6	3) 1.09						
60-65	1	1) 62.3	1) 0.98	1	1) 63.7	1) 1.13	1	1) 60.3	1) 0.76
65-70	3	1) 65.4	1) 0.94	0	-	-	0	-	-
		2) 67.3	2) 0.98						
		3) 69.2	3) 1.07						

APPENDIX Table 4. Parameters used for formation of 2D image for Power 32 kW and substrate temperature 100 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
15-20	1	1) 18.6	1) 1.02	2	1) 16.3 2) 18.4	1) 0.98 2) 1.14	0	-	-
20-25	1	1) 22.3	1) 1.13	1	1) 24.6	1) 1.04	2	1) 20.8 2) 22.6	1) 0.98 2) 1.08
25-30	4	1) 25.6 2) 26.1 3) 27.4 4) 29.8	1) 0.98 2) 1.13 3) 1.04 4) 1.02	4	1) 28.9 2) 27.4 3) 25.1 4) 26.8	1) 1.02 2) 1.13 3) 1.23 4) 1.17	8	1) 26.2 2) 27.4 3) 27.9 4) 28.4 5) 26.2 6) 29.5 7) 28.8 8) 27.6	1) 1.17 2) 1.19 3) 1.01 4) 1.12 5) 1.20 6) 1.13 7) 1.08 8) 1.22
30-35	3	1) 33.6 2) 34.8 3) 30.6	1) 1.09 2) 1.33 3) 0.97	7	1) 34.9 2) 31.8 3) 30.8 4) 30.2 5) 31.7 6) 32.9 7) 33.2	1) 1.02 2) 1.31 3) 0.98 4) 0.86 5) 0.91 6) 0.94 7) 1.13	4	1) 33.6 2) 32.8 3) 34.8 4) 33.5	1) 0.87 2) 0.79 3) 1.03 4) 1.13
35-40	4	1) 35.6 2) 36.8 3) 38.1 4) 39.5	1) 1.13 2) 1.17 3) 1.07 4) 1.09	7	1) 39.6 2) 39.3 3) 38.4 4) 35.1 5) 36.8 6) 38.1	1) 1.02 2) 1.08 3) 0.93 4) 0.92 5) 1.12 6) 1.14	8	1) 35.6 2) 37.9 3) 38.6 4) 35.3 5) 38.4 6) 36.1	1) 1.02 2) 1.01 3) 0.98 4) 0.97 5) 1.13 6) 1.24

					7) 37.5	7) 0.89		7) 38.8	7) 0.88
								8) 39.4	8) 0.91
40-45	1	1) 44.3	1) 1.04	2	1) 42.4	1) 0.92	8	1) 40.6	1) 0.98
					2) 44.8	2) 1.02		2) 41.4	2) 0.87
								3) 43.4	3) 0.79
								4) 44.8	4) 1.12
								5) 40.4	5) 1.32
								6) 40.1	6) 1.14
								7) 41.9	7) 1.11
								8) 42.3	8) 1.09
45-50	2	1) 46.7	1) 0.9	1	1) 47.8	1) 1.15	0	-	-
		2) 47.3	2) 0.97						
50-55	1	1) 50.3	1) 0.93	2	1) 54.3	1) 1.07	3	1) 52.8	1) 0.8
					2) 53.7	2) 0.98		2) 54.9	2) 0.91
								3) 51.4	3) 1.12
55-60	1	1) 57.6	1) 1.21	1	1) 55.6	1) 1.04	0	-	-
60-65	0	-	-	1	1) 63.4	1) 1.13	0	-	-
65-70	0	-	-	1	1) 67.1	1) 0.93	0	-	-

APPENDIX Table 5. Parameters used for formation of 2D image for Power 32 kW and substrate temperature 180 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
15-20	1	1) 19.2	1) 1.03	0	-	-	0	-	-
20-25	4	1) 21.4	1) 1.12	3	1) 20.1	1) 1.14	2	1) 24.6	1) 0.88
		2) 22.3	2) 0.97		2) 21.3	2) 1.04		2) 23.9	2) 0.94
		3) 24.7	3) 0.81		3) 22.4	3) 0.84			
		4) 24.1	4) 0.79						
25-30	5	1) 25.2	1) 0.89	6	1) 27.8	1) 1.12	2	1) 25.1	1) 0.96
		2) 26.7	2) 0.79		2) 28.4	2) 1.03		2) 29.9	2) 0.89
		3) 27.1	3) 0.99		3) 29.8	3) 1.04			
		4) 28.9	4) 1.02		4) 25.2	4) 1.16			
		5) 29.4	5) 1.23		5) 26.3	5) 1.18			
					6) 28.3	6) 0.93			
30-35	2	1) 30.2	1) 1.09	4	1) 34.5	1) 0.87	2	1) 33.4	1) 1.14
		2) 33.4	2) 0.96		2) 33.8	2) 1.07		2) 34.2	2) 1.09
					3) 31.8	3) 1.15			
					4) 30.5	4) 0.95			
35-40	4	1) 38.5	1) 1.01	3	1) 37.8	1) 0.99	3	1) 36.3	1) 1.09
		2) 39.6	2) 0.98		2) 38.9	2) 1.02		2) 37.9	2) 1.17
		3) 35.1	3) 0.95		3) 39.2	3) 1.26		3) 38.1	3) 1.03
		4) 36.3	4) 1.13						
40-45	11	1) 40.1	1) 0.98	3	1) 43.2	1) 1.15	1	1) 43.2	1) 1.02
		2) 41.3	2) 1.12		2) 44.8	2) 1.06			
		3) 41.8	3) 1.14		3) 40.4	3) 1.11			
		4) 42.3	4) 1.19						
		5) 43.5	5) 1.21						
		6) 44.9	6) 1.17						

		7) 42.5	7) 0.89						
		8) 40.9	8) 0.97						
		9) 40.4	9) 0.99						
		10) 41.5	10) 1.4						
		11) 42.8	11) 1.2						
45-50	3	1) 45.9	1) 1.03	2	1) 48.1	1) 1.21	1	1) 48.1	1) 0.98
		2) 46.4	2) 0.98		2) 49.5	2) 1.01			
		3) 49.4							
50-55	2	1) 52.3	1) 0.96	0	-	-	0	-	-
		2) 54.8	2) 0.89						
55-60	1	1) 57.2	1) 0.91	1	1) 58.9	1) 0.91	1	1) 59.1	1) 0.94
60-65	1	1) 63.7	1) 0.96	2	1) 64.9	1) 1.11	1	1) 63.1	1) 1.03
65-70	1	1) 65.9	1) 0.84	1	1) 67.4	1) 1.01	2	1) 68.3	1) 0.98
								2) 69.4	2) 0.83
70-75	2	1) 72.4	1) 0.94	1	1) 73.2	1) 0.81	0	-	-
		2) 74.9	2) 1.13						
75-80	1	1) 78.1	1) 1.03	1	1) 77.9	1) 1.18	0	-	-

APPENDIX Table 6. Parameters used for formation of 2D image for Power 32 kW and substrate temperature 250 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
20-25	1	1) 23.6	1) 0.98	2	1) 23.6 2) 24.7	1) 0.86 2) 1.04	1	1) 20.3	1) 1.12
25-30	5	1) 26.7 2) 27.4 3) 28.9 4) 29.1 5) 26.8	1) 0.88 2) 0.87 3) 0.93 4) 0.96 5) 1.12	2	1) 28.1 2) 29.4	1) 1.08 2) 0.94	2	1) 29.9	1) 1.01
30-35	5	1) 33.4 2) 32.8 3) 34.1 4) 30.8 5) 31.2	1) 1.17 2) 1.03 3) 1.15 4) 1.01 5) 0.96	4	1) 33.6 2) 37.4 3) 39.1 4) 30.8	1) 1.03 2) 1.19 3) 1.02 4) 0.98	2	1) 37.9 2) 38.2	1) 1.03 2) 0.98
35-40	4	1) 36.8 2) 37.2 3) 38.4 4) 39.1	1) 0.98 2) 1.07 3) 1.12 4) 1.21	1	1) 38.9	1) 0.87	3	1) 37.2 2) 35.6 3) 38.9	1) 0.97 2) 1.12 3) 1.03
40-45	5	1) 44.1 2) 40.8 3) 42.6 4) 44.2 5) 43.5	1) 1.11 2) 1.09 3) 1.10 4) 0.98 5) 1.21	6	1) 44.2 2) 43.8 3) 41.8 4) 44.6 5) 42.1	1) 1.01 2) 1.26 3) 0.98 4) 0.89 5) 0.96	1	1) 44.2	1) 0.91
45-50	6	1) 45.1 2) 46.8 3) 47.5	1) 1.01 2) 1.03 3) 1.11	2	1) 46.2 2) 47.8	1) 0.95 2) 1.01	1	1) 48.9	1) 0.88

		4) 49.5	4) 0.99						
		5) 47.3	5) 0.89						
		6) 46.3	6) 0.93						
50-55	7	1) 50.3	1) 1.04	0	-	-	0	-	-
		2) 51.4	2) 1.03						
		3) 54.3	3) 0.93						
		4) 54.8	4) 0.96						
		5) 52.3	5) 0.86						
		6) 54.7	6) 0.89						
		7) 52.8	7) 1.19						
55-60	4	1) 55.7	1) 1.01	1	1) 55.8	1) 1.11	1	1) 58.4	1) 1.08
		2) 58.9	2) 1.24						
		3) 59.8	3) 0.88						
		4) 56.1	4) 0.95						
60-65	1	1) 62.5	1) 0.93	2	1) 64.8	1) 0.93	1	1) 64.9	1.04
					2) 62.3	2) 0.97			
65-70	1	1) 67.9	1) 0.99	1	1) 68.9	1) 1.01	1	1) 69.5	1.21
70-75	2	1) 73.8	1) 1.22	0	-	-	0	-	-
		2) 74.7	2) 1.02						
75-80	1	1) 78.9	1) 0.97	0	-	-	0	-	-
80-85	1	1) 84.6	1) 0.94	1	1) 83.2	1) 1.06	0	-	-

APPENDIX Table 7. Parameters used for formation of 2D image for Power 35 kW and substrate temperature 100 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
20-25	1	1) 22.7	1) 1.08	2	1) 21.3	1) 1.12	3	1) 24.5	1) 1.03
					2) 20.8	2) 0.96		2) 23.7	2) 1.28
								3) 23.8	3) 1.18
25-30	2	1) 25.6	1) 1.09	3	1) 26.7	1) 1.05	2	1) 28.6	1) 1.03
		2) 27.4	2) 1.12		2) 28.4	2) 1.15		2) 25.4	2) 0.98
					3) 29.3	3) 1.20			
30-35	3	1) 30.5	1) 0.98	5	1) 33.2	1) 1.04	2	1) 34.2	1) 0.89
		2) 34.2	2) 1.06		2) 32.4	2) 1.13		2) 33.7	2) 0.76
		3) 33.7	3) 0.87		3) 30.4	3) 1.07			
					4) 34.7	4) 0.99			
					5) 31.9	5) 0.86			
35-40	4	1) 38.9	1) 0.89	3	1) 38.9	1) 0.96	3	1) 38.1	1) 0.94
		2) 35.1	2) 0.94		2) 37.4	2) 0.99		2) 37.1	2) 1.02
		3) 36.3	3) 1.09		3) 38.9	3) 1.02		3) 39.2	3) 1.14
		4) 37.8	4) 1.22						
40-45	1	1) 43.1	1) 1.02	6	1) 41.3	1) 1.02	1	1) 43.2	1) 1.01
					2) 42.4	2) 1.13			
					3) 44.7	3) 1.07			
					4) 44.9	4) 1.2			
					5) 40.1	5) 0.99			
					6) 41.5	6) 0.97			
45-50	2	1) 45.1	1) 1.02	4	1) 47.3	1) 1.02	6	1) 48.1	1) 1.03
		2) 46.8	2) 1.13		2) 49.8	2) 1.13		2) 46.3	2) 1.13
					3) 46.1	3) 0.98		3) 45.9	3) 1.20

					4) 47.4	4) 1.07		4) 46.4 5) 48.1 6) 49.3	4) 0.98 5) 1.14 6) 0.87
50-55	3	1) 50.4 2) 51.6 3) 53.7	1) 1.01 2) 0.94 3) 0.82	4	1) 54.1 2) 53.7 3) 52.6 4) 50.9	1) 1.12 2) 1.09 3) 0.97 4) 1.14	3	1) 51.4 2) 53.8 3) 54.1	1) 0.89 2) 0.91 3) 1.21
55-60	3	1) 57.8 2) 58.6 3) 59.9	1) 1.09 2) 1.19 3) 0.91	0	-	-	0	-	-
60-65	2	1) 60.2 2) 64.8	1) 0.94 2) 0.99	2	1) 63.2 2) 61.5	1) 0.92 2) 0.87	1	1) 64.8	1) 1.32
65-70	1	1) 69.2	1) 1.21	3	1) 67.1 2) 65.2 3) 69.4	1) 0.92 2) 1.21 3) 0.98	1	1) 68.3	1) 1.13
70-75	0	-	-	1	1) 74.3	1) 1.13	0	-	-
75-80	1	1) 77.4	1) 1.22	0	-	-	0	-	-
80-85	2	1) 80.4 2) 81.6	1) 1.13 2) 1.24	0	-	-	0	-	-
85-90	1	1) 88.1	1) 1.08	2	1) 88.6 2) 87.4	1) 0.98 2) 1.02	3	1) 85.1 2) 86.8 3) 88.9	1) 1.13 2) 1.34 3) 0.98

APPENDIX Table 8. Parameters used for formation of 2D image for Power 35 kW and substrate temperature 180 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
20-25	1	1) 18.6	1) 1.21	2	1) 17.3 2) 16.8	1) 1.04 2) 0.97	2	1) 15.3 2) 19.8	1) 0.87 2) 1.13
25-30	3	1) 22.3 2) 23.8 3) 24.7	1) 1.11 2) 1.09 3) 0.98	1	1) 24.8	1) 0.95	1	1) 22.6	1) 1.03
30-35	6	1) 28.9 2) 26.4 3) 25.9 4) 25.1 5) 27.9 6) 29.8	1) 1.01 2) 1.03 3) 0.99 4) 1.00 5) 1.03 6) 1.07	2	1) 28.9 2) 29.5	1) 0.98 2) 1.06	2	1) 27.6 2) 25.2	1) 1.07 2) 1.12
35-40	6	1) 30.3 2) 33.9 3) 34.7 4) 35.1 5) 39.4 6) 39.2	1) 1.01 2) 1.23 3) 1.01 4) 1.00 5) 1.06 6) 0.99	1	1) 33.5	1) 0.98	2	1) 32.6 2) 37.9	1) 1.12 2) 1.02
40-45	4	1) 36.2 2) 35.4 3) 38.9 4) 39.1	1) 1.08 2) 0.95 3) 1.11 4) 0.98	2	1) 39.2	1) 1.03	2	1) 36.4 2) 37.1	1) 1.14 2) 1.04
45-50	4	1) 40.2 2) 41.7 3) 42.9	1) 1.01 2) 0.98 3) 0.97	2	1) 43.9	1) 1.12	2	1) 41.6 2) 42.3	1) 1.04 2) 1.13

		4) 43.6	4) 1.04						
50-55	6	1) 45.8 2) 46.7 3) 47.9 4) 48.3 5) 49.2 6) 49.8	1) 1.01 2) 0.98 3) 1.12 4) 1.13 5) 1.06 6) 1.09	1	1) 48.7	1) 1.02	5	1) 45.1 2) 47.8 3) 48.9 4) 49.2 5) 49.4	1) 1.03 2) 1.14 3) 0.99 4) 0.98 5) 1.01
55-60	2	1) 50.5 2) 53.8	1) 1.12 2) 1.03	1	1) 54.8	1) 1.14	4	1) 54.6 2) 52.5 3) 51.8 4) 50.7	1) 1.04 2) 1.24 3) 0.98 4) 0.87
60-65	2	1) 55.8 2) 59.6	1) 1.09 2) 1.12	1	1) 59.8	1) 1.13	1	1) 59.2	1) 0.95
65-70	2	1) 63.8 2) 64.9	1) 1.22 2) 1.01	0	-	-	0	-	-
70-75	1	1) 68.9	1) 0.98	1	1) 69.4	1) 0.89	2	1) 66.8 2) 67.4	1) 1.22 2) 1.03
75-80	1	1) 73.6	1) 1.03	0	-	-	2	1) 74.8 2) 73.2	1) 1.13 2) 1.09
80-85	2	1) 79.5 2) 78.1	1) 1.01 2) 1.03	0	-	-	1	1) 79.2	1) 1.01
85-90	1	1) 83.2	1) 0.99	2	1) 82.4 2) 81.6	1) 0.98 2) 0.93	2	1) 83.6 2) 84.7	1) 0.97 2) 1.05

APPENDIX Table 9. Parameters used for formation of 2D image for Power 35 kW and substrate temperature 250 °C

Size distribution (μm)	Disk-shaped splats			Fingered (Splashed) splats			Fragmented splats		
	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b	Numbers of splats	Diameter (μm)	a/b
20-25	1	1) 23.5	1) 0.98	2	1) 24.8 2) 23.1	1) 1.02 2) 1.22	1	1) 23.6	1) 1.01
25-30	2	1) 25.2 2) 26.7	1) 1.12 2) 1.13	1	1) 28.9 2) 27.6	1) 1.02 2) 1.13	0	-	-
30-35	3	1) 33.2 2) 34.6 3) 34.9	1) 1.02 2) 0.99 3) 1.03	0	-	-	0	-	-
35-40	2	1) 37.9 2) 38.9	1) 0.95 2) 0.87	1	1) 36.9	1) 1.13	1	1) 37.4	1) 1.22
40-45	8	1) 40.2 2) 41.4 3) 41.6 4) 42.8 5) 43.2 6) 44.6 7) 44.5 8) 44.9	1) 0.88 2) 0.89 3) 1.12 4) 1.01 5) 1.09 6) 1.15 7) 1.09 8) 1.04	2	1) 43.2 2) 44.1	1) 1.01 2) 1.05	2	1) 42.8 2) 44.6	1) 1.04 2) 1.13
45-50	7	1) 45.2 2) 46.8 3) 47.2 4) 48.3 5) 49.2 6) 49.4 7) 49.9	1) 1.22 2) 1.23 3) 0.99 4) 0.98 5) 0.87 6) 0.88 7) 0.78	2	1) 48.3 2) 47.4	1) 1.02 2) 1.03	1	1) 49.2	1) 1.23

50-55	10	1) 50.3 2) 50.8 3) 51.2 4) 52.3 5) 53.4 6) 53.8 7) 54.1 8) 54.3 9) 54.4 10) 54.7	1) 1.01 2) 1.03 3) 1.06 4) 1.08 5) 1.09 6) 0.99 7) 0.87 8) 0.92 9) 0.96 10) 1.0	1	1) 54.3	1) 1.05	0	-	-
55-60	4	1) 55.6 2) 56.9 3) 57.2 4) 59.2	1) 1.03 2) 1.22 3) 1.07 4) 0.88	8	1) 55.1 2) 55.4 3) 55.8 4) 56.5 5) 57.9 6) 58.4 7) 59.4 8) 59.6	1) 0.99 2) 1.21 3) 1.04 4) 1.01 5) 1.10 6) 1.12 7) 1.14 8) 1.02	1	1) 57.4	1) 0.82
60-65	2	1) 63.1 2) 64.7	1) 0.98 2) 1.03	2	1) 60.4 2) 64.9	1) 1.07 2) 1.09	1	1) 64.6	1) 0.97
65-70	2	1) 65.6 2) 66.3	1) 1.01 2) 0.98	1	1) 67.2	1) 1.01	0	-	-
70-75	3	1) 70.3 2) 72.8 3) 74.6	1) 1.04 2) 1.12 3) 0.98	0	-	-	0	-	-
75-80	3	1) 75.8 2) 76.4 3) 78.5	1) 0.99 2) 1.21 3) 0.98	0	-	-	0	-	-
80-85	1	1) 82.8	1) 1.03	1	1) 84.7	1) 1.23	0	-	-

85-90	2	1) 87.4 2) 88.3	1) 0.99 2) 1.13	2	1) 86.4 2) 85.3	1) 1.19 2) 1.07	0	-	-
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