Investigation of the Effects of Particle Temperature and Spacing on Multi-Particle Impacts in Cold Spray

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ABSTRACT

Cold spray deposition is a process in which micrometer sized metal particles are shot upon a surface using a high pressure and temperature gas to form a dense coating. The high strain rates and temperatures occurring in this process significantly affect the material properties of the particles such as flow stress, elastic modulus and thermal properties. A comprehensive understanding of the thermal conditions of both particle and substrate is paramount when assessing the effectiveness of particle deposition for a given input gas condition. Momentum and heat transfer between the particle and gas must be determined through a gas-particle flow model. The heat transfer between the incoming gas and the substrate must be determined as well. In addition to thermal interactions between the gas and material, localized heating occurs during particle plastic deformation. The generated heat may or may not have time to fully dissipate into the substrate before subsequent impacts occur. Therefore, the current state of an initially deposited particle must be accounted for when impacting subsequent particles. In this work, an analytical three-dimensional finite element model utilizing material damage is introduced. A series of single particle impacts followed by two simultaneous impacts are simulated systematically by using conditions common to cold spray procedures. Both aluminum and copper are used for the particles and substrate. This model is then extended to impacting 100 successive particles to form a small coating. Computational results are compared to scanning electron micrographs of coatings created through the cold spray process. In particular, particle morphology is compared for results with similar input parameters. It is found that higher input gas temperatures result in higher particle velocities and temperatures. Furthermore, particles impact with greater kinetic energy and cause more damage to the surface. Copper particles deform more than aluminum particles due to their higher density and thus higher kinetic energy. Accounting for an elevated substrate temperature results in larger crater depth and substrate deformation. As operating temperature increases, the difference in deformation with and without allowing the initial particle to cool becomes more pronounced and simulated deformed particles show similar morphology to those seen in experimental results.
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CHAPTER 1. INTRODUCTION AND LITERATURE SEARCH

1.1 Introduction

This work has two goals:

- Investigate the impacts of multiple particles upon a flat substrate with thermal and momentum variations.
- Quantitatively compare the morphologies of multiple particle impacts using the finite element method with those found using experimental procedures. With this comparison one can determine the validity of the finite element analyses.

In what follows will be a literature search on the current condition of the finite element analysis of particle impacts upon a substrate as it pertains to cold spray technology. An overview of the cold spray process is given in Section 1.2, while the material and coating properties are further explained in Section 1.3. The method of depositing particles is examined in Section 1.4 with the bonding mechanism elucidated in Section 1.5. Section 1.6 explains how the particle temperature and velocity prior to impact can be determined by assessing the gas and particle flow dynamics through the cold spray nozzle. The state of the substrate during particle deposition can have a great effect upon the coating quality and is clarified in Section 1.7. Finally, an overview of the finite element analysis of high velocity particle impacts on a substrate is given in Section 1.8. With this information, a finite element model can be properly developed to study the influences of particle velocity and temperature as well as substrate temperature on the particle deformation.
1.2 Cold Spray Process

Cold particle gas spray, commonly known as cold spray, is a coating process involving high velocity impacts of micron-scale metal particles (10-50 µm diameter) upon a metal substrate. The particles are accelerated in a supersonic gas flow through a converging-diverging de Laval nozzle. High pressure nitrogen or helium gas is used to propel the particles through the nozzle. These particles impact the substrate at 300-1200 m/s and form a layer of metallic coating [1]. This technology was first developed in the 1980’s by a group of scientists at the Institute of Theoretical and Applied Mechanics in Novosibirsk, Russia [2].

Detailed description of a typical de Laval nozzle is given by Alkhimov et al. [3] and presented in Figure 1. Room temperature particles enter the nozzle prior to the converging section and interact with the high pressure, high temperature gas. As the nozzle converges, the gas heats the particles. When the nozzle diverges, the gas accelerates and decreases in temperature due to expansion. The particles accelerate through the diverging section of the nozzle while roughly maintaining their elevated temperatures. Upon exiting the nozzle, the particles and gas travel a short distance before interacting with the substrate.

![Figure 1: Schematic of a cold spray system [4].](image-url)
Cold spray differs from other thermal spray processes in a number of ways, as outlined by Fauchais and Montavon [4]. In thermal spray processes, particles are either partially or fully molten when impacting upon the substrate. These molten particles use thermal energy to bond to the substrate surface. In cold spray, the particles are consistently below their melting temperature and rely on kinetic energy to bond to the surface. The cold spray process offers multiple advantages over other thermal spray processes [5]. The lower particle temperatures prevent unwanted chemical reactions such as oxidation to occur in the particles during flight. In addition, any microstructural properties in the initial powder (ultra-fine grains, specific alloy composition, etc.) are preserved after impact. Klinkov et al. investigated impact procedures of varying magnitude and compared them to impacts derived from the cold spray process, specifically the coefficient of restitution, probability of adhesion, rebound velocity, and bond strength [6]. The similarities between cold spray and explosive compaction were explored by Borchers et al. [7].

1.3 Coating Properties

Typically ductile materials such as aluminum and copper are used as cold spray powder. Ductile metals undergo severe plastic deformation upon impact, forming a smooth coating with little to no porosity and good bond strength [8, 9]. An example of a deposited particle is shown in Figure 2. Trinchi et al. developed a simulation based on empirical data to predict the porosity of a given coating [10]. Ceramics and other hard materials do not exhibit the necessary deformability and instead crack upon impact. The ability of the ductile materials to form relatively strong bonding can be used in a number of applications ranging from filling in minor gaps to building free form
objects [11, 12]. The coatings made through the cold spray process are generally thin (100 µm – 5 cm).

![SEM Image](image)

**Figure 2: SEM of copper particle impacting upon a copper substrate [13].**

The microstructure of cold sprayed coatings can be studied through a scanning electron microscope (SEM). Particle morphology as well as porosity and substrate deformation are clearly shown using this method as seen in Figure 3. When the particle impacts the substrate, it undergoes severe plastic deformation and flattening. Wu et al. measured the flattening ratio (the ratio of deformed height to the diameter of a particle with identical area) of experimentally deposited Al-Si particles upon a steel substrate using SEM and laser scan microscopy (LSM) [14]. They found that flattening ratio is largely independent of the velocity with which the particle impacts the substrate. King and Jahedi investigated the effect of particle size on the deformation upon impact and found that it had a pronounced effect, with larger particles having a larger flattening ratio [15].
1.3.1 Material Properties

Apart from examination of the coating microstructure through SEM, a number of testing procedures can be performed to determine the mechanical properties of coatings, such as elastic/plastic properties, bond strength, electrical resistivity, thermal expansion, etc. [16-20]. Bond strength of cold sprayed coatings was measured for a number of substrate and particle materials in [11, 21]. It was found that the coating elastic modulus was slightly lower than that of the bulk material, suggesting that imperfect metallurgical bonding took place between the particles and substrate. At higher gas temperatures, local annealing occurs in the particles, reducing the internal stresses and decreasing the coating hardness [22]. The existence of internal stresses in the cold sprayed coating is of great importance. High tensile residual stresses can lead to cracking and delamination of the coating. Using neutron diffraction, Luzin et al. developed a
model to determine the residual stresses in aluminum coatings [23]. They divided the stress components into a mechanical and a thermal expansion component. If the coating and substrate are of different materials, the dissimilar thermal strains can produce residual stresses. For particles and substrates made of the same material, Luzin found that small compressive stresses existed in both the substrate and particles.

Bakshi et al. found that the gas used in the cold spray procedure had an effect on the strength of the coating as well, through use of a three point bending test [24]. Higher gas temperatures and pressures lead to higher impact velocities, where the impact velocity has a great effect on the deposition of the particles. Guetta et al. found that differing impact velocities led to variations in the bond strength of the coatings [25].

1.4 Deposition of Particles

As mentioned, the velocity of the impacting particles greatly affects the ability of the particles to adhere to the substrate. Schmidt et al. described in detail how the gas pressure and temperature determine the particle impact velocity and temperature [26]. At low velocities, the particles do not have enough kinetic energy to bond to the substrate. In this velocity regime, the particles bounce off the substrate surface and cause the effects of shot peening [27]. At high velocities, the particles possess excessive kinetic energy and are unable to adhere to the surface. These particles instead erode the substrate [28]. In the middle velocity region, the particles have a high chance of adhesion. In this velocity range, the rebound energy is overcome by the particle-substrate adhesion energy [29-31]. Papyrin et al. compared the rebound and erosion effects
during the cold spray process [32]. Figure 4 highlights the relationship between particle velocity and deposition efficiency of the particles [26].

![Figure 4: Relationship between particle velocity and deposition efficiency [26].](image)

1.4.1 Deposition Efficiency

Although not all particles adhere to the substrate, a high percentage of bonded particles is highly desirable and can be characterized by the deposition efficiency. This deposition efficiency, which can be quantified as the ratio of the number of impacts to the number of bonded particles, can be affected by a number of parameters [33]. Stoltenhoff et al. study the effect of input gas pressure and temperature on the deposition efficiency of copper particles numerically and
experimentally [34, 35]. Li et al. developed a theoretical model to predict the deposition efficiency based on the material spray angle [36, 37]. The highest deposition efficiency occurs during normal impacts, with decreasing efficiency as angle increases. Helfritch and Champagne investigated the effect of particle size on the deposition efficiency of copper particles [38]. Apart from the input parameters, the standoff height of the nozzle away from the substrate has an effect on the deposition efficiency as well [39, 40]. This effect is influenced by the shock region produced by the high velocity gas interacting with the substrate. Samareh and Dolatabadi numerically studied the relationship between particle flow density, standoff height, and shock region size [41]. Li and Li used a stationary nozzle to study the evolution of deposition efficiency of titanium particles at a single location [42].

1.4.2 Critical Velocity

It is generally accepted that in order to have high deposition efficiency, a minimum critical velocity must be reached. Above this critical velocity, the particles possess sufficient kinetic energy to bond to the substrate. This critical velocity has been measured using camera [43, 44] and laser [45, 46] imaging systems. In addition, numerical calculations have been performed to predict the critical velocity of particles under a number of conditions [9, 47, 48]. The critical velocity depends on a multitude of parameters, most importantly the particle size, material density, and material stiffness. For metal particles ranging in diameter of 5-50 µm, this critical velocity was measured to be in the range 300-700 m/s.
1.5 Bonding Mechanism

While it is known that particles, upon surpassing the critical velocity, can impact and adhere to the substrate, the mechanisms with which the particles bond is largely unknown. Multiple hypotheses exist on the nature of particle bonding. Hussain et al. suggest that the bonding is largely due to mechanical interlocking, where the substrate physically entraps the particles [49]. At high impact velocities, the outer region of the particle that impacts the substrate experiences severe plastic deformation and as a result a large temperature increase. It is thought that this temperature spike results in local melting of the particle, where the melted material forms a strong bond with the substrate [50, 51]. The predominant theory is based upon the onset of adiabatic shear instability [13, 52-56]. At high strain rates, heat generated due to plastic deformation is assumed to behave adiabatically and is trapped locally. This local heat generation results in thermal softening of the material and thus further plastic deformation. The material then locally loses its stability and flows as if it were a fluid. Stress-strain curves of a material undergoing deformation with various thermal assumptions are shown in Figure 5.
This flow breaks the oxide layer of the particles and substrate, exposing fresh material to the surface. With the high pressures associated with high velocity impacts, the exposed material forms a high quality metallurgical bond. Furthermore, the critical velocity is thought to be the minimal velocity where adiabatic shear instability occurs along the particle-substrate interface. Dykhuizen et al. investigated the effect of particle diameter on flattening ratio and crater depth numerically to better understand the bonding mechanism [57]. Bae et al. studied the adiabatic shear instability based bonding mechanism numerically by impacting many particles onto a substrate and comparing the results to experimental findings [58]. Champagne et al. looked at material mixing between copper particle and aluminum substrates as a result of adiabatic shear instability as well [59].
1.6 Gas and Particle Dynamics

Now that it is established that a critical particle velocity must be reached in order to obtain successful deposition, significant work has been done to model the fluid dynamics within the nozzle. Computational Fluid Dynamics (CFD) has been performed to determine the particle and gas velocities and temperatures throughout the nozzle. Initially, one-dimensional computations were performed to determine the gas-particle flow relationship throughout the particle flight as well as particle conditions prior to impact [34, 35, 60-62]. These simulations assumed the one-dimensional fluid flow was isentropic and adiabatic. In addition, two-dimensional axisymmetric models were simulated [63-68]. Figure 6 shows the gas and particle velocity and temperature throughout the nozzle and into the substrate obtained by Particle FlowSim, a software package developed by our group.

![Figure 6: Particle and gas velocity and temperature throughout the cold spray nozzle.](image)

Figure 6: Particle and gas velocity and temperature throughout the cold spray nozzle.
Upon developing the computational model to calculate the particle flow through the nozzle, the effects of varying input parameters can be researched. Helfritch and Champagne developed a fluid flow model to characterize the effect of particle diameter on impact velocity and deposition efficiency [38]. Takana et al. investigated the assistance of electrostatic forces on the impact behavior of nano- and micro-scaled particles, finding that the electrostatic forces helped propel particles previously obstructed by the shock wave [69, 70]. Karimi et al. used a three-dimensional model to study the variation of particle velocity with respect to location within the nozzle at the nozzle exit and found that particles located near the center of the nozzle exited with a higher velocity than those located near the nozzle edge [71].

By performing CFD, the particle velocity and temperature of a given input system can be predicted. This data can then be used in conjunction with accepted critical velocity data to predict whether the particles have a good chance of adhesion. In addition, the nozzle itself can be designed and optimized to maximize particle velocity [61, 72-76]. Assadi et al. developed a series of parameters to predict particle impact velocities based upon the initial input conditions [77]. Samareh and Dolatabadi performed simulations for a full three-dimensional model to determine the optimal nozzle position and substrate shape as well as the effect of particle flow density [41, 78].

1.7 Substrate Temperature

Though the majority of research has been focused on the particle conditions prior to and during impact, recent research has shown that for the initial layer of particles, the state of the substrate has a great effect as well [79]. For subsequent layers beyond the initial layer, the impacting
particles bond to the previously deposited particles and have little interaction with the substrate. Higher deposition efficiency as a result of elevated substrate temperatures was studied by Fukumoto et al. [80]. Higher substrate temperatures resulted in greater substrate deformation and a higher chance of activating the shear instability mechanism. Fukumoto et al. found that, even for particles at room temperature, the deposition efficiency was higher for hot substrates than it was for room temperature substrates. Yu et al. numerically investigated the effect of a preheated substrate on the flattening ratio of copper particles [81]. McDonald et al., Yin et al. and Kosarev et al. numerically calculated the effect of the input gas on the temperature of the substrate and found that the substrate temperature was a function of the radial distance away from the gas spray point [82-85]. For small substrates, the size of the substrate has an effect on the fluid flow and particle impact velocity [86]. In general, the substrate is large enough that this effect is negligible.

1.8 Finite Element Analysis

In addition to extensive experimentation, finite element analyses (FEA) of particle impacts have been performed. Due to the short time span and small sizes of individual impacts, FEA provides an excellent opportunity to monitor the stress and temperature states of the particle and substrate throughout the impact process. Other thermal spray processes, such as high velocity oxygen fueled spray (HVOF), have been studied using the finite element method by Zhan et al. [87] and Bansal et al. [88]. Extensive research of the cold spray process has been performed by using the finite element method as well. Upon simulating the particle impacts, the results can be used to compare to experimental results under similar input conditions. The particle morphology can be
directly compared to those obtained from experiments. Characteristics such as crater depth and flattening ratio can be quantified to validate the accuracy of the simulations [57, 89].

The adiabatic shear instability can be observed in finite element simulations as well. Strain levels along the particle-substrate interface are monitored throughout the simulation. Prediction of a spike in strain is interpreted as the onset of shear instability, shown in Figure 7 [13, 52, 90]. In addition, particle rebound velocity can be measured to determine the amount of energy lost during impact [91]. Low rebound energy can be overcome by higher adhesion energies and lead to bonding. The existence of a critical velocity has been confirmed using the finite element method in this fashion [13, 37, 48].

![Figure 7: Time history of strain at a point on the particle interface for different velocities. Adiabatic shear instability occurs above 550 m/s [13].](image)
1.8.1 Model Properties

In order to properly set up a finite element model, many parameters such as material model, dimensions, and possible failure criteria must be selected. While many material models exist for large deformation of metals, a material model developed by Johnson and Cook has been able to accurately predict large deformations at high strain rates [92]. This model takes into account the temperature softening and strain rate hardening effects found during deformation. Because temperature and strain rate effects are immensely important in cold spray impacts, the model developed by Johnson and Cook has been widely used to simulate particle impacts [9, 13, 25, 37, 47, 48, 52, 55, 56, 58, 91, 93-102]. Dislocation mechanics-based models such as Zerilli-Armstrong and Steinberg-Guinan-Lund are less common but have been accurately used as well [54, 57, 103]. Figure 8 shows the stress strain curve for a typical elastic-plastic model and the Johnson Cook material model [104].

![Stress strain curves for elastic-plastic and Johnson Cook models](image)

Figure 8: Stress strain curves for elastic-plastic and Johnson Cook models [104].
1.8.2 Model Size Effects

The size of the model has an influence on the simulation results. Schmidt et al. investigated the effect of particle size on the critical velocity and then compared the cold spray impacts with other high velocity phenomenon such as explosive compaction and ballistic impacts [47]. Li et al. studied the effect of mesh size as well as the implementation of a number of controls to improve the finite element model results [56, 102]. A change in mesh size resulted in slightly different temperature and strain values along the particle-substrate interface. Implementing a material damage parameter helped improve the overall quality of the deformation with the exception of at the interface. Yildirim et al. expanded upon this work and developed a material damage model to prevent excessive distortion of the particles [105]. The addition of distortion control greatly improved the level of deformation found in the particle.

1.8.3 Deformation Control

Another method of controlling excessive distortion is to utilize Arbitrary Lagrangian Eulerian mechanics (ALE). The addition of ALE refined the deformation near the particle-substrate interface and produced results that are closer to those found in experiments [102]. Using ALE reduces the effect of the element size used in the simulations and allows for more complex tests. Eulerian based models have been used to simulate particle impacts and accurately predict particle critical velocities and other characteristics [98, 101]. Modeling the particles using smoothed particle hydrodynamics (SPH) has also been studied [106-108].

The dimensionality of the simulation has an effect as well. Two-dimensional simulations, typically with axial symmetry, provide a low cost method of determining single particle impacts.
For multiple particle impacts as well as oblique impacts, a computationally expensive three-dimensional model must be used. As with the two-dimensional models, the Johnson Cook material model has been typically used [89, 90, 107, 109]. Grujicic et al. used the Steinberg-Guinan-Lund model to simulate many particle impacts as a means to study the viability of depositing polymers to metal substrates [110].

1.8.4 Effect of Substrate

As mentioned previously, the substrate condition can affect the deposition ability of impacting particles [100]. The temperature of the substrate can lead to a difference in crater size for a given particle impact according to Yu et al. [81]. Kumar et al. investigated the effect of a roughened substrate where a particle impacts upon either a mound or crater that is smaller, larger, or of similar size to the impacting particle [97]. They also studied the contact time and area as a function of the impact velocity upon the rough substrate and determined that these parameters play a significant role in the adhesion ability of impacting particles. The temperature of a substrate plays a major role as well and will be investigated further in this work.

The presence of an outer oxide layer has been investigated by Li et al. in order to determine how it changes the deformation of the particle [95]. They found that the critical velocity of a particle largely depended on the thickness of its oxide layer. Yin et al. concluded that the high velocity impact broke the oxide layer into pieces which exposed new material to the substrate [111].
Apart from the substrate condition, the angle between the incoming particle and the substrate can determine the ability of particles to adhere to the substrate [93, 94]. Larger angles result in a lower velocity normal to the substrate and thus a lower flattening ratio.

1.8.5 Multiple Particles

Recently, particle impacts have been expanded to include multiple particles to attempt to understand bonding between particles rather than just between an initial particle and the substrate. Bae et al. impacted many titanium particles to study the possible presence of the adiabatic shear instability at particle interfaces [58]. Zhou et al. simulated a particle impacting directly on a previously deposited particle as well as 2 secondary particles impacting subsequently upon an initial particle [112]. Yin et al. impacted multiple particles in various configurations to understand how particle interfaces evolved throughout the coating time [99, 101, 107]. Subsequent impacts had a profound effect upon the deformation of the initial particle and could help improve the deposition efficiency.

1.9 Scope of This Work

In this work, the finite element method is utilized to simulate a particle impact followed by two simultaneous secondary impacts. Effects of high-strain rate, plastic heating, heat transfer through the substrate and the particle, and bonding are considered in the models. Research suggests that, due to the low particle density within the gas, impacts occur independently with sufficient time to allow the particles to cool before experiencing further impacts [34]. To simulate this effect, an intermediate cooling step is added between the initial and secondary impacts. This cooling effect lowers the substrate and initial particles to the substrate temperature
before experiencing the subsequent impact. On the other hand the constant stream of warm gas heats the substrate to a superambient temperature. Effects of various modeling assumptions related to substrate and particle temperatures are studied systematically. The substrate is tested at room temperature as well as an elevated temperature that will be determined by a two-dimensional heat transfer analysis, as described later. The particle morphologies which are determined through FEA are compared to SEM micrographs of aluminum and copper coatings obtained with the same system parameters, in order to assess the effects of modeling assumptions.

In order to obtain results similar to an experimental coating, the presented model is extended to impacting 100 successive particles upon a single substrate. Digital image processing is performed to analyze the particle morphologies in both the FEA model and experimental findings. The purpose of this image processing is to attempt to identify and quantify certain morphological characteristics of the deposited particles such as flattening ratio. The image processing is performed for experimental coatings and FEA models for multiple inlet gas temperatures. The FEA models are simulated for both room temperature and heated substrates.
CHAPTER 2. EXPERIMENTS

2.1 Introduction

In this chapter, the experimental setup is presented. Aluminum and copper were deposited on same material substrates by Plasma Giken, Tokyo, Japan. The de Laval nozzle used in this deposition process is described in detail. Finally, the sample preparation method for the SEM investigations is explained.

2.2 Experimental Methods

Aluminum and copper powder particles, shown in Figure 9, were cold sprayed onto aluminum and copper plates. The particle-substrate combinations used in the experiments are designated in the rest of the paper as Cu-Cu, Cu-Al, Al-Cu, and Al-Al, where the first material indicates the particle and the second material indicates the substrate. The aluminum powder particles were spherical in shape with a mean diameter of 31 µm, while the copper particles were amorphously shaped, with a mean diameter of 25 µm.

Figure 9: Representation of the aluminum (left) and copper (right) particles.
These powders were sprayed by using a de Laval type converging-diverging nozzle, with the dimensions shown in Figure 10 and specified in Table 1, in a Plasma Giken cold-spray system (Plasma Giken, Tokyo, Japan). Nitrogen gas, at a pressure of 3 MPa, was used to propel the powder through the nozzle on to the substrate with a standoff distance, $L_{st}$, of 30 mm. Various inlet gas temperatures, $T_G$, ranging from 573-1273 K were used in order to obtain different impact velocities, $V_{i}^{(p)}$, and impact temperatures, $T_{i}^{(p)}$, as listed in Table 2. The impact velocities and temperatures of the particles were computed by a one-dimensional (1D) gas flow model, as explained later.

![Figure 10: Schematic representation of the converging-diverging nozzle used in this work.](image-url)
Table 1: Nozzle dimensions.

<table>
<thead>
<tr>
<th>Name</th>
<th>Symbol</th>
<th>Value (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Diameter of Inlet</td>
<td>(d_0)</td>
<td>15</td>
</tr>
<tr>
<td>Diameter of Throat</td>
<td>(d_{th})</td>
<td>3</td>
</tr>
<tr>
<td>Diameter of Outlet</td>
<td>(d_e)</td>
<td>6.5</td>
</tr>
<tr>
<td>Length of Convergence</td>
<td>(L_{con})</td>
<td>71</td>
</tr>
<tr>
<td>Standoff Length</td>
<td>(L_{st})</td>
<td>30</td>
</tr>
<tr>
<td>Length of Divergence</td>
<td>(L_{div})</td>
<td>181</td>
</tr>
</tbody>
</table>

Table 2: Initial gas temperatures \(T_G\) used in the experiments, and the corresponding temperatures and velocities of the particles computed by using the 1D particle flow simulations. The inter-particle spacing \(\delta\) values used three particle impact simulations.

<table>
<thead>
<tr>
<th>Particle</th>
<th>Substrate</th>
<th>Gas Temp (T_G) (K)</th>
<th>Velocity (V_i^{(p)}) (m/s)</th>
<th>Particle Temp (T_i^{(p)}) (K)</th>
<th>Spacing (\delta) ((\mu)m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>Cu</td>
<td>1273</td>
<td>725</td>
<td>921</td>
<td>10, 20, 30, 40</td>
</tr>
<tr>
<td>Cu</td>
<td>Cu</td>
<td>1073</td>
<td>684</td>
<td>778</td>
<td>10, 20, 30, 40</td>
</tr>
<tr>
<td>Cu</td>
<td>Cu</td>
<td>873</td>
<td>636</td>
<td>635</td>
<td>10, 20, 30, 40</td>
</tr>
<tr>
<td>Cu</td>
<td>Al</td>
<td>1273</td>
<td>725</td>
<td>921</td>
<td>30</td>
</tr>
<tr>
<td>Cu</td>
<td>Al</td>
<td>1073</td>
<td>684</td>
<td>778</td>
<td>30</td>
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<tr>
<td>Cu</td>
<td>Al</td>
<td>873</td>
<td>636</td>
<td>635</td>
<td>30</td>
</tr>
<tr>
<td>Al</td>
<td>Cu</td>
<td>773</td>
<td>677</td>
<td>602</td>
<td>30</td>
</tr>
<tr>
<td>Al</td>
<td>Cu</td>
<td>673</td>
<td>632</td>
<td>525</td>
<td>30</td>
</tr>
<tr>
<td>Al</td>
<td>Cu</td>
<td>573</td>
<td>581</td>
<td>449</td>
<td>30</td>
</tr>
<tr>
<td>Al</td>
<td>Al</td>
<td>773</td>
<td>677</td>
<td>602</td>
<td>10, 20, 30, 40</td>
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<tr>
<td>Al</td>
<td>Al</td>
<td>673</td>
<td>632</td>
<td>525</td>
<td>10, 20, 30, 40</td>
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<tr>
<td>Al</td>
<td>Al</td>
<td>573</td>
<td>581</td>
<td>449</td>
<td>10, 20, 30, 40</td>
</tr>
</tbody>
</table>

A Hitachi S4800 high resolution field-emission scanning electron microscope (SEM) was used to investigate the deformed particle shapes in the coatings. Coated samples were sliced and then polished mechanically with progressively fine sand paper ranging from 240 to 1200 grit. 1 \(\mu\)m
and 0.3 µm polishing paste was used to polish the surface, which was then etched in order to identify the particle boundaries in the coating and the grain boundaries in the particles. The details of the etchants used for each sample are given in Table 3.

<table>
<thead>
<tr>
<th>Coat Material</th>
<th>Etchant</th>
<th>Concentration</th>
<th>Conditions</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>Distilled water</td>
<td>20 ml</td>
<td>10 s</td>
</tr>
<tr>
<td></td>
<td>Hydrochloric acid</td>
<td>5 ml</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Ferric chloride</td>
<td>1 gram</td>
<td></td>
</tr>
<tr>
<td>Al</td>
<td>Distilled water</td>
<td>95 ml</td>
<td>60 s</td>
</tr>
<tr>
<td></td>
<td>Hydrochloric acid</td>
<td>1.5 ml</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Nitric acid</td>
<td>2.5 ml</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Hydrofluoric acid</td>
<td>1 ml</td>
<td></td>
</tr>
</tbody>
</table>

Table 3: Etching conditions used in preparing the samples for SEM imaging and analysis.
CHAPTER 3. THEORY

3.1 Introduction

In this chapter the mechanics behind high strain rate impacts are presented and the finite element model used in this work is described. The material model developed by Johnson and Cook for high strain rate and high temperature deformation is explained in Section 3.2. A material damage model based upon shear failure is described as well. A finite element model is implemented in Section 3.3 where an initial particle impacts upon a flat substrate followed by two secondary particles.

The momentum and thermodynamic relations between the carrier gas, particles, and substrate are explained in Section 3.4. A gas-particle flow model is developed in Section 3.4.1 and used to calculate the particle velocity and temperature prior to impact in Section 3.4.2. The heat transfer between the incoming gas and the substrate is investigated for both a 1D and 2D substrate in Section 3.4.3 and Section 3.4.4, respectively. The substrate temperatures used in this work are calculated in Section 3.4.5. The average time between particle impacts upon the same location is calculated and compared to the cooling rate of particles in Section 3.4.6. A particle may impact upon a recently deposited particle which may not have had time to cool to the substrate temperature completely. In this study, where the temperature of the substrate is varied, the temperature of the initial particle cannot be neglected.
3.2 Material Definition

At high-strain rates the material flow stress $\sigma$ depends on the equivalent plastic strain $\varepsilon$, the strain rate $\dot{\varepsilon}$, and temperature $T$ [92]. In this work the flow stress for high-strain rate plastic deformation was modeled by using the Johnson-Cook plasticity model given as follows [92],

$$\sigma = \left( A + B\varepsilon^n \right) \left( 1 + C \ln \frac{\dot{\varepsilon}}{\dot{\varepsilon}_0} \right) \left[ 1 - \left( \frac{T - T_R}{T_m - T_R} \right)^m \right]$$

(1)

where $\dot{\varepsilon}_0$ is the reference strain rate, $T_R$ is the reference temperature and $T_m$ is the melting temperature of the material. $A$, $B$, $C$, $n$ and $m$ are empirically determined constants, listed in Table 4 [92, 113].

<table>
<thead>
<tr>
<th></th>
<th>OFHC Copper [92]</th>
<th>Aluminum 1100H12 [113]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>90</td>
<td>148.4</td>
</tr>
<tr>
<td>$B$</td>
<td>292</td>
<td>345.5</td>
</tr>
<tr>
<td>$C$</td>
<td>0.025</td>
<td>0.001</td>
</tr>
<tr>
<td>$n$</td>
<td>0.31</td>
<td>0.183</td>
</tr>
<tr>
<td>$m$</td>
<td>1.09</td>
<td>0.859</td>
</tr>
<tr>
<td>$T_m$ (K)</td>
<td>1356</td>
<td>933</td>
</tr>
<tr>
<td>$T_R$ (K)</td>
<td>293</td>
<td>293</td>
</tr>
<tr>
<td>$\rho$ (g/cm$^3$)</td>
<td>Temp Dependent [114]</td>
<td>Temp Dependent [114]</td>
</tr>
<tr>
<td>$E$ (GPa)</td>
<td>Temp Dependent [114]</td>
<td>Temp Dependent [114]</td>
</tr>
<tr>
<td>$\alpha$ (1/K)</td>
<td>Temp Dependent [114]</td>
<td>Temp Dependent [114]</td>
</tr>
<tr>
<td>$k$ (W/m*K)</td>
<td>Temp Dependent [114]</td>
<td>Temp Dependent [114]</td>
</tr>
<tr>
<td>$C_p$ (J/kg*K)</td>
<td>Temp Dependent [114]</td>
<td>Temp Dependent [114]</td>
</tr>
<tr>
<td>$D_p$ (µm)</td>
<td>25</td>
<td>31</td>
</tr>
</tbody>
</table>

Table 4: Material constants for the strain rate dependent flow stress model of Johnson-Cook [92, 112].
In order to alleviate the effects of severe mesh distortion and to properly model the material response, Yildirim et al. used the Lagrangian mechanics with material damage [105]. Both ductile and shear failure can occur in materials undergoing high strain-rate deformation. Ductile failure occurs due to nucleation, growth, and coalescence of voids, whereas shear failure is based on shear band localization. In cold spray of spherical particles material failure primarily occurs due to shear [105]. In this work, material damage was based on a shear failure model. The critical strain value for this damage model was based on adiabatic shear instability. Upon plastic deformation at high strain rates, the material experiences a combination of strain and strain rate hardening as well as localized thermal softening. The thermodynamic energy balance between plastic strain energy and heat generation is given by the following relationship,

$$\int_{T_0}^{T} \rho C_p dT = \beta \int_{\varepsilon_p}^{\varepsilon} \sigma d\varepsilon$$  \hspace{1cm} (2)$$

where $C_p$ is the specific heat constant, $\rho$ is the material density, $T$ and $T_0$ are the temperature and reference temperature, respectively, and $\beta$ is the inelastic heat fraction. The value of $\beta$ was set to 0.9, as it is generally agreed that 90% of the plastic strain energy is dissipated as heat [115]. It was shown in reference [105] that the instability strain approximately equals 2 which is used in this work. The density, $\rho$, elastic modulus, $E$, coefficient of thermal expansion, $\alpha$, and thermal conductivity, $k$, of the materials are listed in Table 4. Note that temperature dependent values are used for both aluminum and copper [114].

Particle attachment to a substrate, or onto another particle, can be due to one of the aforementioned mechanisms. In this work the cohesive particle attachment was modeled. A
theoretical cohesive strength of 350 MPa was used when particles come in contact with each other or the substrate. In general, this cohesive strength should be an arbitrary fraction of the material’s failure stress, where the fractional value accounts for surface defects such as oxidation and impurities during contact. The value used in this work assumes a very high likelihood of particle cohesion. The effects of the cohesive strength as a materials parameter have been reported recently by Yildirim et al. [105].

3.3 Simulations

In this work cold spray of the four particle-substrate material combinations used in the experiments was simulated by the finite element method (FEM). One of the particles, called here the initial particle, impacts a flat substrate normally and is followed by the simultaneous impact of the two secondary particles. In the horizontal direction, the secondary particles were spaced symmetrically about the initial particle by an inter-particle spacing value, $\delta$. The Cu-Cu and Al-Al impacts were simulated with $\delta$ ranging from 10 to 40 $\mu$m, whereas the Cu-Al and Al-Cu simulations were simulated with $\delta = 30$ $\mu$m. In the vertical direction, the secondary particles were placed at an appropriate height above the substrate, in order to allow the initial particle to impact and find steady state before the subsequent impacts occur.

The aforementioned material model is incorporated into a coupled temperature-displacement dynamic simulation using the finite element analysis (FEA) program, Abaqus/Explicit (Simulia Inc. Providence, RI). Figure 11 shows the initial setup for the simulation with $\delta = 30$ $\mu$m between the secondary particles. The particles and the segment of the substrate near the impact region use a mesh size that is $1/25^{th}$ of the particle diameter $D_p$. In order to ensure that the
substrate can be assumed semi-infinite, the contact duration must be shorter than the time for the waves to reflect back from the boundaries to the impact site. This distance was determined by numerical tests. The substrate was constrained on its outer faces for all degrees of freedom. A coarser substrate mesh was used further away from the impact site to limit the number of elements and improve simulation run time.

![Finite element mesh](image1)

Figure 11: The finite element mesh used to model the substrate, the primary and secondary particles. Note that the particle diameter is 25 µm.

### 3.4 The Model of Gas-Particle Interactions and Substrate Heating

In cold spray, particles gain speed in a supersonic gas flow. The supersonic flow is achieved by introducing a compressed, pre-heated gas into a converging-diverging nozzle, shown in Figure 10. Particles are added into the gas flow before the converging section of the nozzle and are
typically at room temperature. The gas velocity increases in the diverging section of the nozzle due to expansion, and the gas temperature rapidly drops from its preheated level. The particles gain substantial speed in this section [61]. In order to maximize particle impact velocity, cold-spray nozzles are designed to keep the gas flow supersonic inside the diverging section. In addition to momentum, the particles also exchange heat with the gas due to forced convection, and their temperatures rise. Outside the nozzle, the gas travels a relatively short distance and experiences a shock before encountering the coated surface, eventually decelerating and spreading over the coated surface. The gas temperature rises in the shock region while its velocity slows down.

Recent work on the effects of substrate temperature showed that increasing the substrate surface temperature increases the particle deformation [116], and it will likely increase the deposition efficiency for some metals [57, 80]. When the high velocity gas impinges upon the substrate, it spreads radially along the substrate surface. The substrate temperature increases relative to the reference, due to convective heat transfer through its surface, while the heat is conducted away into the substrate and along its surface through conductive heat transfer. Kosarev et al., and Ryabinin et al. presented 1D and 2D models, respectively, for the substrate surface temperature [83, 84]. Yin et al. used a commercially available computational fluid dynamics program to analyze the same problem for a number of substrates and found that substrates with low thermal conductivities produce higher surface temperatures as well as larger temperature gradients [85].

In this work a MATLAB based simulation tool was developed to compute the velocity and temperature of the carrier gas and the particles between the nozzle entry point and the substrate,
and the substrate surface temperature, by using the models described next. The particle impact velocity and temperature as well as the substrate surface temperature at the time of impact are found by using this simulation tool and are incorporated into the finite element model of the particle impact.

3.4.1 The Model of Gas-Particle Interactions

The gas velocity, \( V_g \), and temperature, \( T_g \), along the nozzle axis (\( z \)) are found from a 1D gas flow model. For the typical particle feed rate used in cold spray applications the presence of the particles does not significantly affect the main gas flow, therefore the flow is modeled in single-phase. The flow inside the converging-diverging nozzle is modeled as a 1D, isentropic flow [1], and the gas velocity and temperature are assumed to remain constant between the nozzle exit and the shock region near the substrate. In the shock region, the flow conditions are modeled as described by Dinavahi et al. [64]. The governing equations for the 1D compressible flow in the nozzle, and in the shock region are omitted for brevity, but can be easily found in [1, 64].

The particle exchanges momentum with the gas due to fluid drag forces. The particle mechanics along the \( z \)-axis is represented by its equation of motion as follows.

\[
m_p V_p \frac{dV_p}{dz} = \frac{1}{2} C_D A_p \rho_p \left( V_g - V_p \right) \left| V_g - V_p \right|
\]

where, \( V_p \) is the particle velocity, \( m_p \) and \( \rho_p \) are the particle mass and particle density, respectively, \( A_p \) is the cross-sectional area of the particle, and \( C_D \) is the drag coefficient for a sphere [61].
Heat transfer between the gas stream and the particle is found by using the following form of the convective heat balance,

\[ m_p c_p V_p \frac{dT_p}{dz} = h_{gp} A_p \left( T_g - T_p \right) \]  

(4)

where \( T_p \) is the particle temperature, \( c_p \) is the heat capacity of the particle, and \( h_{gp} \) is the convection heat transfer coefficient. The values of the parameters used for the gas flow are given in Table 5 [34, 61, 116].

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Copper</th>
<th>Aluminum</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \rho ) (kg/m(^3))</td>
<td>8,933</td>
<td>2,702</td>
</tr>
<tr>
<td>( m_p ) (kg)</td>
<td>1.248*10(^{-13})</td>
<td>6.545*10(^{-14})</td>
</tr>
<tr>
<td>( A_p ) ((\mu)m(^2))</td>
<td>1,963</td>
<td>3,019</td>
</tr>
<tr>
<td>( c_p ) (J/kg*K)</td>
<td>390</td>
<td>910</td>
</tr>
<tr>
<td>( k ) (W/m*K)</td>
<td>400</td>
<td>220</td>
</tr>
</tbody>
</table>

Table 5: Gas flow parameters.

3.4.2 Calculated Gas-Particle Interaction

Figure 12 shows the variation of the temperature and velocity of the gas and of the particle are computed between the nozzle inlet and the substrate surface, along the nozzle axis (\( z \)-direction). This case is computed for the experimental conditions given in Table 2. The inlet pressure of the nitrogen gas modeled in this example is 3 MPa with the inlet gas temperature as shown on the figure.
Figure 12: Computed variation of the temperature and velocity for the carrier gas ($N_2$) and the particles along the nozzle and the shock region for aluminum (left) and copper (right). Gas flow parameters are given in Table 5.
The particles are introduced into the converging section of the nozzle with zero velocity, at room temperature (293 K). While the gas velocity increases in the converging section of the nozzle, its temperature remains constant. On the other hand, the particle temperature increases to the gas temperature. In the diverging section of the nozzle the gas expands and its velocity continues to increase while its temperature drops. In response, the particle velocity continues to increase while it loses heat to the surrounding colder gas. The flow conditions between the nozzle exit and the substrate are assumed to be constant until the shock region, in which the gas velocity returns to zero and temperature recovers to the $T_G$, as the flow is assumed to be adiabatic. After leaving the nozzle exit, the particle continues to increase its velocity, and lose its heat. However, these are small changes, and the particle impacts the surface with a speed similar to that of the nozzle exit. Table 6 gives $T_i^{(p)}$ for each inlet gas temperature used in these simulations.

<table>
<thead>
<tr>
<th>Particle</th>
<th>Gas Temp $T_G$ (K)</th>
<th>Particle Velocity $V_i^{(p)}$ (m/s)</th>
<th>Particle Temp $T_i^{(p)}$ (K)</th>
<th>Surface Temp $T_s$ (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>1273</td>
<td>725</td>
<td>921</td>
<td>811</td>
</tr>
<tr>
<td>Cu</td>
<td>1073</td>
<td>684</td>
<td>778</td>
<td>705</td>
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<td>Cu</td>
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<td>Al</td>
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<td>567</td>
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<td>Al</td>
<td>673</td>
<td>632</td>
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<td>510</td>
</tr>
<tr>
<td>Al</td>
<td>573</td>
<td>581</td>
<td>449</td>
<td>453</td>
</tr>
</tbody>
</table>

Table 6: Particle and substrate temperatures for each inlet gas temperature.
3.4.3 1D Model of Substrate Heating

The heat exchange between the gas and the substrate is modeled at steady state. Kosarev et al. showed that the 1D heat conduction equation on the substrate surface can be modeled by the following relationship [84],

\[
\frac{\partial^2 T_s}{\partial x^2} = \frac{h_{gs}}{t_s} k \left( T_{gs} - T_s \right) \tag{5}
\]

where \( x \) is the coordinate direction along the surface, \( T_s = T_s(x) \) is the surface temperature of the substrate material, \( T_{gs} = T_{gs}(x) \) is the gas temperature along the surface, \( h_{gs} = h_{gs}(x) \) is the convective heat transfer coefficient, \( k \) is the thermal conductivity of the substrate, \( t_s \) is the substrate thickness, and \( L \) is the substrate length. Thermal conductivities of the modeled materials are given in Table 5.

The conservation of thermal energy along the substrate surface is ensured by the following relationship as described by Kosarev et al. [84],

\[
\int_{0}^{L} h_{gs} \left( T_{gs} - T_s \right) dx = 0 \tag{6}
\]

The center of gas impingement point, located at \( x = 0 \) is a symmetry point, represented by the following boundary condition,

\[
\frac{\partial T_s}{\partial x} \bigg|_{x=0} = 0 \tag{7}
\]
The temperature, $T_{gs}$, and the convective heat transfer coefficient, $h_{gs}$, of a gas impinging upon a flat surface vary significantly along the surface. This variation has been characterized experimentally for cold spray applications [1, 83]. In order to determine the gas temperature along the surface of the substrate, $T_{gs}$, Kosarev et al. present the following relationship,

$$
\bar{T}_{gs}(\bar{x}) = \left(1 + 15\bar{x}^2\right)^{-0.25}
$$

(8)

where $\bar{T}_{gs}(\bar{x}) = (T_{gs} - T_R) / (T^{(g)}_{i} - T_R)$ with $T_R$ as the reference temperature and $T^{(g)}_{i}$ as the gas temperature at the impingement point, and $\bar{x} = x / x_{1/2}$ where $x_{1/2}$ is the half width of the gas pressure profile on the surface [84]. $x_{1/2}$ is approximated as half the nozzle exit diameter, $d_e$, for standoff heights $L_{sf} < 4d_e$. Equations (6 - 8) are solved simultaneously with the $h_{gs}$ and $T_{gs}$ variations computed, as described above, to obtain the substrate temperature along the surface, $T_s(x)$.

### 3.4.4 2D Model of Substrate Heating

The heat exchange between the carrier gas and the substrate can be expressed in 2D as follows [83],

$$
\frac{\partial^2 T_s}{\partial x^2} + \frac{\partial^2 T_s}{\partial y^2} = \frac{h_{gs}}{t_s k} (T_{gs} - T_s)
$$

(9)

where $T_{gs}$, $T_s$, and $h_{gs}$ are now functions of $x$ and $y$. The jet impacts the substrate at the origin, and the flow can be assumed to be symmetric. Only one quarter of the solution domain is
simulated and this spans $0 \leq x \leq L$ and $0 \leq y \leq L$. The symmetry boundary conditions are given as follows,

$$\frac{\partial T}{\partial y} = 0 \text{ at } y = 0, \ 0 \leq x \leq L \quad \text{and} \quad \frac{\partial T}{\partial x} = 0 \text{ at } x = 0, \ 0 \leq y \leq L$$

(10)

It is assumed that no heat transfer takes place along the far edges of the substrate [83]. These insulated edges are expressed mathematically as follows,

$$\frac{\partial T}{\partial y} = 0 \text{ at } y = L, \ 0 \leq x \leq L \quad \text{and} \quad \frac{\partial T}{\partial x} = 0 \text{ at } x = L, \ 0 \leq y \leq L$$

(11)

The gas temperature $T_{gs}$ and the Nusselt number $Nu = \frac{h_{gs}d_e}{k_g}$ along the substrate were experimentally determined and expressed in non-dimensional form by Ryabinin et al. in Figures 2 and 3 of their paper [83]. Note that $k_g$ is the thermal conductivity of the gas in the definition of the Nusselt number. In this work, the gas temperature along the substrate surface $T_{gs}$ and the convective heat transfer coefficient $h_{gs}$ were determined by fitting a curve to the data presented by Ryabinin [83]. Thus $T_{gs}$ and $h_{gs}$ were obtained as a function of the radial distance

$$r = \sqrt{x^2 + y^2}.$$  

Similar to Section 3.4.3, Equations (9-11) were solved numerically to obtain the substrate temperature $T_s(x,y)$ along the substrate surface.

### 3.4.5 Calculated Substrate Heating

Upon determining the gas and particle impingement conditions as described in Section 3.4.3 for each experimental condition, the 2D heat transfer model given in Section 3.4.4 was used to find
the steady state gas temperature, $T_{gs}$, and the substrate temperature, $T_s$, along the substrate surface. Figure 13 shows $T_{gs}$ and $T_s$ in the radial direction for each experimental condition for a 3 mm thick substrate. In general, for substrates with high thermal conductivity ($k \geq 40 \text{ W/(m*K)}$), the surface temperature of the substrate at the point of spraying is significantly lower than $T_i^{(g)}$. For such substrates (e.g. aluminum and copper), redistribution of heat within the substrate substantially decreases the substrate surface temperature below the gas temperature $T_i^{(g)}$. For example, Figure 13 shows that the surface temperature of the substrate, $T_s$, is higher than gas temperature, $T_{gs}$, further away from the impingement point ($x = 0$), due to conduction within the substrate as well as the hot gas mixing with the cool ambient air. Table 6 shows the steady state surface temperature of the substrate at the impingement point based on the results of the model presented above. It is seen that for five of the six cases tested the particle is warmer than the substrate, and in fact the relationship $T_G > T_i^{(p)} > T_s$ is predicted. In the case of the Al particle injected with 573 K gas temperature the substrate is 4 degrees warmer than the particle. In this work the temperature value obtained at $x = 0$ will be used as the substrate temperature in some of the impact simulations presented below.
Figure 13: Computed $T_s$ and $T_{gs}$ for each inlet gas condition.
3.4.6 Cooling Effect

Because particles are heated in the gas flow, it is essential to evaluate the effects of particle temperature during and after impact. Figure 14a and 14b show the temperature histories of single aluminum and copper particles, respectively, based on single particle impact simulations. Each particle is impacted upon a heated (black) and room temperature (red) substrate. Regardless of the substrate temperature or the particle material, each particle approaches the substrate temperature in approximately 5 µs.

![Temperature histories of single aluminum and copper particles](image)

Figure 14: Computed temperature histories of single aluminum (left, \(D_p = 31 \, \mu m, V_i^{(p)} = 677 \, m/s, T_i^{(p)} = 602 \, K\)) and copper (right, \(up = 25 \, \mu m, V_i^{(p)} = 725 \, m/s, T_i^{(p)} = 921 \, K\)) particles impacting upon a cool (red) and hot (black) substrate.

In multi-particle impacts the cooling time and frequency of impact would affect the material properties. An estimate of the time between two consecutive impacts at the same location is next...
developed to assess whether the particle will have sufficient time to cool. For a given feed rate \( f_p \) (mass/time) and particle mass \( m_p \), the total number of impacts per unit time is found as follows, \( N_{\text{impact}} = \frac{f_p}{m_p} \). The cross-sectional area of the nozzle exit, \( A_e \), and the approximate area of a deformed particle, \( A_d \), give the total number of particles making impact on the surface. Then the number of particles impacting a specific location per unit time can approximated as \( \left( \frac{f_p}{m_p} \right) \left( \frac{A_d}{A_e} \right) \). The average time between two consecutive impacts on a specific location, \( t_{\text{ave}} \), can then be estimated as follows,

\[
\begin{align*}
    t_{\text{ave}} &= \left( \frac{m_p}{f_p} \right) \left( \frac{A_e}{A_d} \right) \\
    &= \frac{4 \pi \rho_p \left( \frac{1}{2} d_p \right) ^3}{6 f_p} \frac{A_e}{4 \pi \left( \frac{1}{2} d_p \right) ^3} \\
    &= \frac{\rho_p d_p A_e}{6 f_p}
\end{align*}
\]

(12)

where \( \rho_p \) is the mass density of the particles. Note that the deformed area of the particle is estimated to be 4 times its surface area to obtain this relationship. Using this relationship \( t_{\text{ave}} \), for cold spray of 25 µm diameter copper particles \( (\rho_p = 8933 \text{ kg/m}^3) \) with a feed rate of \( f_p = 0.01 \text{ kg/s} \) through a nozzle with an exit diameter of \( d_e = 6.5 \text{ mm} \), is found as 124 ms. Comparing this value with the cooling time of a single particle upon impact, it is seen that an impacted particle could have sufficient time to cool down to the substrate temperature. For aluminum particles impacting upon a heated substrate, the difference in particle and substrate temperature is small. Heat generation due to plastic deformation, however, increases the temperature of the newly deposited particles well above that of the substrate. In what follows the effects of cooling the impacted particle on the particle deformations will be computed and compared to the case where the impacted particle is hit by subsequent particles before it cools down. In the latter case both hot and cold substrates will be simulated.
CHAPTER 4. RESULTS

4.1 Introduction

The goal of this chapter is to investigate the effects of the (assumed) thermal state of the particles on the finite element simulations of the particle deformations. The effects of multiple impacts are also explored by a systematic study of impact of three particles. To this end, the particle velocity and temperature and the substrate temperature related to the conditions used in the experiments are investigated. Each parameter combination is simulated with and without allowing the initial particle time to cool before being subsequently impacted. Both copper and aluminum are simulated. Section 4.2 examines the particle morphology in experimental results and highlights some characteristics that may be found in analytical results. Section 4.3 displays the results of all of the simulations. Section 4.3.1 and Section 4.3.2 show results for impacts upon a room temperature substrate with and without allowing the initial particle to cool, respectively. These findings are then compared in Section 4.3.3. Section 4.3.4 and Section 4.3.5 show impacts upon a heated substrate (Table 6) with and without allowing the initial particle to cool and are compared in Section 4.3.6. Section 4.3.7 compares all of the simulations based upon substrate temperature. The presence of residual compressive stresses is examined in Section 4.3.8.

4.2 Experimentally Observed Particle Morphologies in the Coating

Figure 15 shows SEM images of cold sprayed coatings of Al-Al and Cu-Cu, respectively. These samples were produced with inlet gas temperatures $T_G$ of 573 K and 1273 K, respectively. While the exact velocities and temperatures upon impact were not measured, by using the flow model
presented above the impact velocities were estimated to be 581 m/s and 725 m/s for the aluminum and copper samples, respectively (Table 2).

Figure 15: SEM of Al-Al at $T_G = 573$ K (left) and Cu-Cu at $T_G = 1273$ K (right) gas temperature.

Boundaries between the deposited particles, as well as some voids present in the sample are clearly seen in the SEM images of the aluminum sample. The variation in particle-size observed in Figure 15a is attributed to the fact that this cross-section does not necessarily represent the geometric center of each particle, and also to the size variation in the original powder of particles. Flattened edges are observed at most particle-particle interfaces, whereas curved edges are observed where voids are present. Interfacial jets and self-interlocking are not apparent in these particle-particle interfaces despite being known to occur in particle-substrate interfaces [13]. The largely indirect impacts of particles onto the uneven partially coated surface may allow particles to undergo significant deformation without creating a visible interfacial jet. The box in Figure 15a highlights two particles that may have indirectly impacted upon a center particle equally and will be compared to the simulation results later in the paper.
It is more difficult to see the inter-particle boundaries in the SEM images of the copper particle Figure 15b. In general, copper particles appear to be experiencing much larger deformation than aluminum particles, resulting in flatter particles and less inter-particle voids. The etchant used for the sample preparation affected the inter-particle grain boundaries, as well as the particle-to-particle boundaries in a similar way. Consequently, it is difficult to identify individual particles. It should also be noted that the original particles were amorphous contributing to the difficulty of boundary identification. With this said, one can also argue, based on the observation that particle and grain boundaries were etched in a similar manner, that significant amount of metallurgical bonding took place in this coating and that the particle boundaries behaved as grain boundaries.

4.3 Simulated Impacts

Impact of three particles was simulated for different material combinations, particle velocities, substrate temperatures, and inter-particle separation distance $\delta$ values. Most of the papers in the related literature neglect the effects of the initial particle and substrate temperatures on deformation, and assume these to make contact at room temperature. On the other hand, as discussed above, the most likely scenario includes the particle temperature determined by using a gas flow analysis and the substrate temperature determined by analyzing the substrate heating due to the heat transfer from the carrier gas. Effects of various modeling assumptions are presented in this paper. First, the substrate is kept at the room temperature of 293 K and the effects of particle cooling (or lack thereof) are investigated. The purpose of this cooling is to study the effect of initial particle temperature during the deformation of secondary particles. The cooling effect simulates a particle that has sufficient time to cool prior to secondary impact while
the lack of cooling simulates secondary particles that impact nearly instantaneously after the initial impact. Next, the substrate is kept at the elevated steady state temperature value found as described in Section 3.4.5. The effects of cooling the particle to the substrate temperature (or lack thereof) are investigated for this condition as well. The calculated temperature conditions are summarized in Table 6.

4.3.1 Impacts Conditions: Cool Substrate and No-Cooling of the Primary Particle

Figure 16 shows the results of the impact of aluminum particles for impact velocities of $V_{i}^{(p)} = 581, 632$ and $677$ m/s. The corresponding values of the initial particle temperatures are $T_{i}^{(p)} = 449, 525$ and $602$ K, respectively, shown in Table 6. The initial separation distance between the secondary particles are $\delta = 10, 20, 30$ and $40$ µm. These figures show the final morphologies and the equivalent strain distributions in the particles and the substrate. In addition to impacting upon aluminum substrates, particles are also impacted upon a copper substrate with $\delta = 30$ µm.
Al-Al Impacts without cooling and $T_s = 293$ K

$V_{i}^{(p)} = 581$ m/s, $T_{i}^{(p)} = 449$ K: $V_{i}^{(p)} = 632$ m/s, $T_{i}^{(p)} = 525$ K: $V_{i}^{(p)} = 677$ m/s, $T_{i}^{(p)} = 602$ K

$\delta =$

40

$\mu$

$m$

30

$\delta =$

$\mu$

$m$

Al-Cu Impacts without cooling and $T_s = 293$ K

$V_{i}^{(p)} = 581$ m/s, $T_{i}^{(p)} = 449$ K: $V_{i}^{(p)} = 632$ m/s, $T_{i}^{(p)} = 525$ K: $V_{i}^{(p)} = 677$ m/s, $T_{i}^{(p)} = 602$ K

Figure 16: Equivalent strain distributions and the final particle morphologies for the aluminum particles where the secondary particles impact the primary particle immediately after its impact with $T_s = 293$ K.

Deformation of the initial particle increases with increasing impact velocity and decreasing inter-particle separation. As $\delta$ increases, and the initial particle experiences less direct impact, it experiences lower distortion and damage. As with the particle-particle interfaces of aluminum presented in the SEM image (Figure 15a), the particle-particle interfaces in the simulations also exhibit relatively “flat” contact surfaces. Interfacial jets occur for some $(V_{i}^{(p)}, \delta)$ combinations between the secondary particles and the substrate. The highest strains and most damage occur along the particle boundaries, due to large shear strain values. This effect becomes more pronounced as the inter-particle separation increases. When comparing the aluminum and copper substrates, one can see that the particles deform slightly more when impacting a copper...
substrate. The higher strength and density of the copper produces a stiffer substrate into which the particles impact, causing slightly increased deformation.

Impact of copper particles with impact velocities of $V_i = 636, 684$ and $725 \text{ m/s}$ are presented in Figure 17. These velocities correspond to initial particle impact temperatures of $T_i = 635, 778$ and $921 \text{ K}$, respectively, as given in Table 6. As with the impacts in Figure 16, a series of impacts on an aluminum substrate with $\delta = 30 \mu\text{m}$ is simulated to highlight the difference in substrate material upon the particle deformation.

![Cu-Cu Impacts without cooling and $T_s = 293 \text{ K}$](image)

![Cu-Al Impacts without cooling and $T_s = 293 \text{ K}$](image)

Figure 17: Equivalent strain distributions and the final particle morphologies for the copper particles where the secondary particles impact the primary particle immediately after its impact with $T_s = 293 \text{ K}$. 
As with the aluminum, particle distortion increases with increasing velocity and temperature, and with decreasing inter particle separation. However, the copper particles experience significantly more deformation than the aluminum particles. Because of the higher gas temperatures used during these procedures, copper particles impact at higher temperatures and velocities. This combined with higher density of copper results in more kinetic energy at the point of impact. The stronger impact causes the particles to undergo severe strains and as a result significant material damage. For small $\delta$, the initial particle is completely deleted at the center cross section. The impacts of copper particles upon an aluminum substrate create a larger crater than those impacting upon a copper substrate. The larger crater depth is due to the fact that an aluminum substrate is softer than a copper substrate.

4.3.2 Impacts Conditions: Cool Substrate with Cooled Primary Particle

In order to study the effect of initial particle temperature on the deformation of secondary particles, an intermediate cooling step must be added. The cooling simulates that the particle has had enough time to reach equilibrium with the substrate prior to the secondary impacts. Figure 18 shows the case where the primary particle is allowed to cool to the substrate temperature of 293 K before the impact of the secondary particles.
Comparison with Figure 16 shows a lower amount of distortion of the initial particles. As the initial particle impact temperature increases, the difference between the cooled and non-cooled cases becomes more pronounced. This difference is attributed to the inherent temperature dependence of the Johnson-Cook plasticity model. The particle morphologies of the aluminum impacts, particularly Figure 18g and Figure 18j, match up reasonably well with the SEM image presented in Figure 15a. While the particles shown in Figure 15a experience further deformation due to the particles impacting afterward, they still exhibit similar morphology as the three particles shown in these FE predictions.
The implementation of particle cooling is extended to simulations using copper particles as well. Figure 19 shows impacts of copper particles onto a cool substrate after the initial particle had enough time to cool to room temperature.

Figure 19: Equivalent strain distributions and the final particle morphologies for the copper particles where the secondary particles impact the primary after it cools down to room temperature with $T_s = 293$ K.

Because cold spray procedures depositing copper particles operate at higher temperatures, the addition of an intermediate cooling has a more pronounced effect. Though the initial particle still experiences significant deformation and element deletion, it is less damaged than the
simulations without the cooling step included shown in Figure 17. While the deformation in Figure 19 seems too severe at elevated temperatures, the results for the lower temperature simulations match well with experimental results. The initial particle in Figure 19f shows a strong similarity to the conical shaped particle highlighted in Figure 15b.

4.3.3 Comparison of Simulations with Cool Substrate

The addition of the cooling effect increases the stiffness in the initial particle and reduces particle deformation. As temperature increases, the difference between the initial particle deformation with and without the cooling effect grows. Very little difference is seen at the lowest operating temperatures while the highest temperatures show a significant difference. These comparisons suggest that, due to the average time between impacts, the addition of the intermediate cooling step is crucial to properly representing the multi-particle impact conditions present in cold spray processes when assuming a cold substrate.

4.3.4 Impacts Conditions: Heated Substrate and No-Cooling of the Primary Particle

While the previous simulations used a substrate temperature of 293 K, it has been shown that heat transfer between the gas and substrate results in the substrate temperature, $T_s$, to rise above 293 K. The values of $T_s$ are given in Table 6. Figure 20 shows the results for aluminum particles impacting upon an aluminum substrate with an elevated temperature. No intermediate cooling step is applied to these simulations.
Figure 20: Equivalent strain distributions and the final particle morphologies for the aluminum particles where the secondary particles impact the primary particle immediately after its impact with a heated substrate.

In comparison to Figure 16, the particles form larger craters within the substrate. Due to the temperature dependence of the Johnson-Cook material model, the hotter substrate is softer than a substrate at room temperature. As a result, the initial particle experiences less deformation. As with the previous simulations, increasing $\delta$ results in decreasing deformation in the initial particle.

Figure 21 shows the impacts of copper particles upon a heated copper substrate. As with the hot aluminum substrate, these impacts result in a larger crater depth. For higher gas temperatures and small $\delta$, the initial particle is almost completely deleted. In addition, the secondary particles create a massive crater and experience extensive deformation as well. The largely flattened particles closely resemble those found in certain regions of Figure 15b. Note that deletion of the...
elements representing a substantially large volume of material (particle) represents a severe deformation. In the modeling approach used in this work, the elements are removed from analysis. Severely deformed material in actual experiments would be expected to remain in the impact area but perhaps flow and spread more readily. A different modeling approach that deals with severe deformation without element deletion is required to predict the fate of such elements.

Figure 21: Equivalent strain distributions and the final particle morphologies for the copper particles where the secondary particles impact the primary particle immediately after its impact with a heated substrate.

4.3.5 Impacts Conditions: Heated Substrate and Cooling of the Primary Particle

Figure 22 shows the results for aluminum particles and substrate with an intermediate cooling step added. In this cooling step, the initial particle and substrate are reset to the initial substrate
temperature, $T_s$. Comparing Figure 20 to Figure 22 shows that less deformation occurs in the initial particle when the cooling step is applied, which is consistent with the simulations involving a room temperature substrate in Figure 16 and Figure 18.

Finally, simulations involving copper particles upon a heated substrate with intermediate cooling are presented in Figure 23. These results are largely similar to the results in Figure 21 with only a slight difference in the deformation of the particles. As with previous simulations of copper particles, the initial particle is nearly completely deleted for small $\delta$. 

Figure 22: Equivalent strain distributions and the final particle morphologies for the aluminum particle where the secondary particles impact the primary after it cools down to the heated substrate temperature.
4.3.6 Comparison of Simulations with Heated Substrate

While the addition of a cooling step had a pronounced effect for the simulations involving a room temperature substrate, the effect was less noticeable for simulations with a heated substrate. When the substrate and initial particle are allowed time to cool, they reach the hotter temperature provided by the incoming gas. This change is far less than the difference between a hot particle and 293 K substrate, reducing the effect. As a result, the particle morphologies in the simulations where the initial particle was allowed time to cool are fairly similar to the morphologies in the simulations with near instantaneous impacts.
4.3.7 Comparison of Substrate Temperatures

In general, the initial particle deformations with a higher temperature substrate are less severe than the deformations with the substrate temperature of 293 K. In the warm and cold substrate simulations without the cooling step, the initial particle is near the particle impact temperature $T_i^{(p)}$. During the warm substrate simulations, the warmer substrate takes some of the deformation energy from the initial particle, resulting in less particle deformation and a deeper crater depth.

When the initial particle and substrate are allowed to cool to the initial substrate temperature of 293 K, the initial particle temperature drops and produces the stiffest particle with the least deformation. The most deformation is seen in the simulations involving a warm substrate where the initial particle did not have time to cool. The differences in these simulations are mainly attributed the relative temperature of the initial particle, where the majority of deformation occurs. For the hot substrate results where the particle had time to cool, the initial particle is reset to substrate temperature, $T_s$, which is below the particle temperature, $T_i^{(p)}$, but above the room temperature 293 K. The middle particle temperature, shown in Figure 20 and Figure 21, produces particle deformation that is more than the cold substrate with the cooling step but less deformation than the warm substrate without a cooling step applied. These results show that the effect of substrate temperature and cooling of the initial particle must be accounted for when modeling multiple particle impacts.

4.3.8 Residual Stresses

It has been shown that the presence of residual compressive stresses in the cold spray coating can be beneficial to the coating strength. In order to investigate the presence of residual compressive
stresses in these finite element analyses, the minimum principal stress was monitored for a simulation that is representative of a multiple particle impact. This simulation, with the final plastic equivalent strain profile highlighted in Figure 20i, had a particle velocity and temperature of $V_i^{(p)} = 677$ m/s and $T_i^{(p)} = 602$ K, respectively, with a heated substrate temperature of $T_s = 567$ K. Figure 24 shows the time history of the minimum principal stress at various points during the simulation.

![Image of stress distribution](image)

Figure 24: Time history of minimum principal stress with $V_i^{(p)} = 677$ m/s, $T_i^{(p)} = 602$ K and $T_s = 567$ K.

Prior to the secondary impact, the substrate shows a residual compressive stress while the particle experiences a smaller amount of residual stress. During the secondary impact, the primary and secondary particles show significantly larger compressive stresses, as expected.
Shortly after the impact, however, a tensile stress emerges in the center of the primary particle. This tensile stress could be a result of the particle attempting to rebound after compressing. As the particles approach a steady stress state, they show a similar stress profile as the initially deposited particle in Figure 24a. The residual stress may depend on the temperature and velocity with which the particles impact the substrate. Figure 25 shows the minimum principal stress state momentarily before secondary impact upon a heated substrate for each input gas condition in Table 2 for inter-particle width of $\delta = 30 \, \mu\text{m}$.

$$V_i^{(p)} = 581 \, \text{m/s}, \ T_i^{(p)} = 449 \, \text{K} \quad T_s = 453 \, \text{K};$$
$$V_i^{(p)} = 632 \, \text{m/s}, \ T_i^{(p)} = 525 \, \text{K} \quad T_s = 510 \, \text{K};$$
$$V_i^{(p)} = 677 \, \text{m/s}, \ T_i^{(p)} = 602 \, \text{K} \quad T_s = 567 \, \text{K};$$

Figure 25: Minimum principal stress for each input gas condition in Table 2 for $\delta = 30 \, \mu\text{m}$ momentarily before secondary impact.

As particle temperature and velocity decrease, larger compressive stresses are seen in the initial particle and substrate. The larger stresses are most likely due to the colder and stiffer material resisting deformation. Figure 26 shows the minimum principal stress state after the secondary particles impact upon the substrate.
Al-Al Impacts without cooling and a heated substrate

\[ V_{i}^{(p)} = 581 \text{ m/s}, \quad T_{i}^{(p)} = 449 \text{ K}, \quad T_s = 453 \text{ K}; \]
\[ V_{i}^{(p)} = 632 \text{ m/s}, \quad T_{i}^{(p)} = 525 \text{ K}, \quad T_s = 510 \text{ K}; \]
\[ V_{i}^{(p)} = 677 \text{ m/s}, \quad T_{i}^{(p)} = 602 \text{ K}, \quad T_s = 567 \text{ K}; \]

Some small tensile stresses (gray contour color) are seen in the particles at low temperatures and inter-particle width \( \delta \). These stresses may be present because the particles are attempting to rebound from the surface but are held in place by the interfacial cohesion. As particle temperature and velocity increase, these tensile stresses decrease. Smaller compressive stresses are seen in the substrates for higher temperature and velocity impacts as well. Over time, a deposited coating would most likely show compressive stresses in the deposited particles with a higher compressive stress in the substrate. These compressive stresses agree with what has been seen experimentally and would be beneficial to a cold sprayed coating [23].

Figure 26: Minimum principal stress for aluminum particles impacting upon a heated substrate immediately after initial impact.
CHAPTER 5. CHARACTERIZATION OF DEFORMED PARTICLE MORPHOLOGIES IN COLD GAS PARTICLE SPRAY

5.1 Introduction

Although the analyses presented in Chapter 3, involving impact of 3 particles upon a flat substrate, help illustrate the effects of particle-particle interactions, they do not fully capture the effects of depositing a full coating of material. In order to better compare numerical and experimental results, this model is extended to impacting 100 successive particles. Comparisons of the experiments and the simulations are carried out by identifying characteristic traits of the deformed particle shapes by image processing in MATLAB. The 100 particle impact finite element model is produced in Section 5.2.1 with results presented in Section 5.2.2. The image processing method is described in Section 5.3. Section 5.4.1 and Section 5.4.2 show the image processing results for 100 particle impacts upon a room temperature and heated substrate, respectively, for each impact condition in Table 6. Section 5.5 displays the results of image processing for SEM’s of particle coatings made with each inlet gas condition. Finally, Section 5.6 compares the data for each image processing parameter.

5.2.1 100 Particle Impact

The simulations described in Chapter 3 involved 3 particles impacting on a flat substrate. Realistically, many particles impact upon previously deposited particle layers. In order to better understand the particle-particle interactions, the finite element analysis was extended to impact of 100 successive particles.
The plastic deformation once again is modeled using the Johnson-Cook plasticity model given in Equation (1) with experimentally determined constants listed in Table 4. Lagrangian mechanics with material damage was used to determine element damage and failure. This failure was based upon the shear failure model presented by Yildirim et al. [105]. The critical strain value of 2 was based upon the adiabatic shear instability, determined by the combination of heat generation due to plastic deformation and material thermal softening. The density, \( \rho \), elastic modulus, \( E \), coefficient of thermal expansion, \( \alpha \), and thermal conductivity, \( k \), of aluminum are listed in Table 4. Temperature dependent values are used for some parameters.

Cohesive attachment between the particles and the substrate was included in these simulations. Rather than using a fraction of the yield stress as a limit for cohesive particle attachment, perfect cohesive behavior was used. This ensured that all particles remained attached to the substrate once they made contact, and did not interfere with the incoming particles. Because copper particles show relatively large deformation upon impact, only aluminum particles were used. If copper particles were simulated, subsequent particle impacts would almost completely eliminate previous particles. Each particle had a diameter of 31 \( \mu \text{m} \). The particles and region of the substrate near the particle impact area used a mesh size that was \( 1/10^{\text{th}} \) of the particle diameter \( D_p \). A coarser mesh was used further away from the impact area. The impact positions of the particles were randomly selected on the horizontal \((x, y)\) plane as shown in Figure 27.
All particles had equal spacing in the vertical ($z$) direction and positioned to impact shortly after the previous particle. Particles impacted at the temperature and velocity listed in Table 6. Heat transfer between the particles and substrate slowly cooled the particles to a temperature approaching the substrate temperature. However, the impacts occurred shortly after one another, leaving little time for the recently deposited particle to reach the substrate temperature. The implementation of an artificial cooling step was not studied as this would increase simulation times. Therefore, these simulations model the effect of closely impacting particles rather than particles that have sufficient time between impacts to cool. This implies a high “flow rate”. Both hot and room temperature substrates were simulated.
5.2.2 100 Particle Results

Figure 28 shows an isoparametric view of the deposited particles upon the substrate. All particles deposited within an area of $10D_p \times 10D_p$.

![Figure 28: Deposited aluminum particles in 100 particle simulation.](image)

Figure 29 shows the equivalent plastic strain and particle morphologies for each input condition given in Table 6 for room temperature and heated substrates. All simulations show a cross section at the center of the model perpendicular to the y-axis,
Figure 29: Equivalent strain distributions for 100 particle impacts of each of the inlet gas conditions given in Table 6 with room temperature (left) and heated (right) substrates.

As particle temperature and velocity increase, the particle deformation greatly increases. Previous particles at the highest inlet temperature are nearly destroyed due to the further deformation caused by subsequent impacts. As expected, the heated substrate experiences more deformation than the room temperature substrate. With the exception of the initially impacted particles, the morphology of the particles between hot and cold substrates is very similar. This shows that, regardless of substrate temperature, the particles are mainly affected by the most
recent layer of particles. Figure 30 shows the von Mises stress distributions for each input condition in Table 6. The von Mises stress can be used to understand the overall elastic stress state of the material.

Figure 30: Von Mises stress distributions for 100 particle impacts of each of the inlet gas conditions given in Table 6 with room temperature (left) and heated (right) substrates.
Higher residual stresses are seen in the room temperature substrates. With the exception of the first layer of particles, similar stresses are seen throughout the particle layers. The lower residual stress in the substrate may be due to the softer material.

5.3 Materials and Methods

Numerous parameters are used to quantify the particle morphologies at various inlet gas conditions. MATLAB provides many commands under the group `regionprops` that help characterize digital images. The following shape identification metrics have been used in this work.

**Axis ratio**: is the ratio of the major and minor axes of an ellipse that is fit to particle given shape. The major and minor axes are calculated by creating an ellipse that has the same normalized second central moments as the original object.

**Eccentricity**: is the ratio of the distance between the foci of the ellipse and the length of its major axis. An object with an eccentricity value of 0 is a perfect circle while a value of 1 corresponds to a line.

**Orientation angle**: is the angle of the major axis with respect to the horizontal plane, given in degrees.

**Equivalent diameter**: is the diameter of a circle that has the same area as the object.

**Perimeter**: is the actual length of the perimeter of the object.
In addition to the functions available in MATLAB, other morphological characteristics are defined by Mikli et al. [117]. Among these parameters, the following are used.

**Elongation:** similar to the axis ratio, it is defined as,

\[ EL = \log_2 \left( \frac{a}{b} \right) \]  

where \( a \) and \( b \) are the major and minor axis lengths as used before, respectively. An example of various shapes with differing elongation is given in Figure 31.

![Figure 31: Samples with different values of elongation [117].](image)

**Dispersion:** is the measure of edge smoothness of the object. Dispersion of a perfect ellipse is zero while it grows with increasing roughness of the perimeter, as shown in Figure 32. Dispersion is defined mathematically as follows,

\[ DP = \log_2 (\pi ab) \]
Roundness: is the measure of the circularity of the object. A perfect circle has roundness value of 1 and a line has a value approaching infinity. Roundness is defined as,

\[
RN = \frac{P^2}{4\pi A}
\]  

(15)

\(A\) is the area of the object and \(P\) is the perimeter. Roundness is similar to the eccentricity defined earlier. Equivalent diameter and perimeter are in units of pixels and will be converted to \(\mu\)m later. Orientation angle is in degrees and all other parameters are non-dimensional.

5.4.1 Image Processing of Cold Substrate

These metrics will be applied to both the numerical results of the 100 particle impacts as well as images of experimental results under similar input conditions. The simulations presented in Figure 29 can be analyzed by using these parameters. Multiple images are obtained from each
simulation by taking cross sectional cuts spaced 10 µm apart in both the x- and y-directions.

These cuts range from -110 µm to +110 µm with respect to the center of the model, giving a total of 46 images. Figure 33 shows a cut at the center of the model (distance = 0) for both the x- and y-directions.

Figure 33: Image cuts perpendicular to the x (left) and y (right) axes for 100 aluminum particle impacts ($D_p = 31$ µm, $V_i^{(p)} = 581$ m/s, $T_i^{(p)} = 449$ K).

A MATLAB macro is written to carry out the following image processing tasks. Each cross section image is transferred to a black and white image. After this transfer, objects below a certain size threshold are removed. These particles were assumed to be small edge pieces away from the geometric center and misrepresentative of the entire particle morphology. This threshold allows only particles with sufficiently large cross sections to be analyzed, removing smaller end pieces that may distort the data. The number of particles found in each image ranges from 1-28 objects and totals 804 objects across the 46 images. Figure 34 shows a black and white image of the objects above the threshold value for cut at the center of the model perpendicular to the y-axis, taken from Figure 33b.
Particle shapes in each image are analyzed by using the metrics introduced previously. The resulting data for each metric is then taken from each image cut of the simulation results and compiled into a series of histograms. Figure 35 through Figure 40 show the results of aluminum impacts for each input condition in Table 6.
Figure 35: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 581$ m/s, $T_i^{(p)} = 449$ K and $T_s = 293$ K.
Figure 36: Perimeter, Elongation, Dispersion and Roundness histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 581 \text{ m/s}, T_i^{(p)} = 449 \text{ K} \text{ and } T_s = 293 \text{ K}.$
Figure 37: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 632$ m/s, $T_i^{(p)} = 525$ K and $T_s = 293$ K.
Figure 38: Perimeter, Elongation, Dispersion and Roundness histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 632$ m/s, $T_i^{(p)} = 525$ K and $T_s = 293$ K.
Figure 39: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for 100 particle impacts of aluminum with $V_i(p) = 677$ m/s, $T_i(p) = 602$ K and $T_s = 293$ K.
Figure 40: Perimeter, Elongation, Dispersion and Roundness histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 677$ m/s, $T_i^{(p)} = 602$ K and $T_s = 293$ K.

Table 7 shows the averages and standard deviations of all parameters in Figure 35 through Figure 40. A preliminary measurement of an undeformed particle showed that a 31 µm particle spans 129 pixels, or 3.69 pixels/µm. This conversion factor was used for calculating the equivalent diameter and perimeter in µm.
<table>
<thead>
<tr>
<th>Sample, Temperature (K)</th>
<th>$V_i^{(p)} = 581$ m/s</th>
<th>$V_i^{(p)} = 632$ m/s</th>
<th>$V_i^{(p)} = 677$ m/s</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_i^{(p)} = 449$ K</td>
<td>$T_i^{(p)} = 525$ K</td>
<td>$T_i^{(p)} = 602$ K</td>
<td></td>
</tr>
<tr>
<td><strong>FEA Characteristic</strong></td>
<td><strong>Average</strong></td>
<td><strong>Std Dev</strong></td>
<td><strong>Average</strong></td>
</tr>
<tr>
<td>Axis Ratio</td>
<td>2.77</td>
<td>1.23</td>
<td>3.72</td>
</tr>
<tr>
<td>Eccentricity</td>
<td>0.89</td>
<td>0.08</td>
<td>0.93</td>
</tr>
<tr>
<td>Equivalent Diameter (pixels)</td>
<td>79.11</td>
<td>27.08</td>
<td>64.01</td>
</tr>
<tr>
<td>Equivalent Diameter (µm)</td>
<td>21.44</td>
<td>7.34</td>
<td>17.35</td>
</tr>
<tr>
<td>Orientation Angle (degrees)</td>
<td>-1.18</td>
<td>26.01</td>
<td>-2.18</td>
</tr>
<tr>
<td>Perimeter (pixels)</td>
<td>378.09</td>
<td>131.68</td>
<td>344.79</td>
</tr>
<tr>
<td>Perimeter (µm)</td>
<td>102.46</td>
<td>35.68</td>
<td>93.44</td>
</tr>
<tr>
<td>Elongation</td>
<td>1.36</td>
<td>0.55</td>
<td>1.71</td>
</tr>
<tr>
<td>Dispersion</td>
<td>0.15</td>
<td>0.13</td>
<td>0.19</td>
</tr>
<tr>
<td>Roundness</td>
<td>2.51</td>
<td>1.12</td>
<td>3.28</td>
</tr>
</tbody>
</table>

Table 7: Average and standard deviations of image processing parameters for each numerical simulation analyzed in Figure 35 through Figure 40.

The simulation corresponding to $V_i^{(p)} = 581$ m/s and $T_i^{(p)} = 449$ K ($T_G = 573$ K) has a lower axis ratio and elongation than the other two simulations. The higher temperature results in particles that are more flattened. For the $V_i^{(p)} = 677$ m/s and $T_i^{(p)} = 600$ K ($T_G = 773$ K) case, subsequently impacted particles may be flattened so much that they are nearly completely deleted, leaving behind only less damaged particles. The eccentricity for each simulation has roughly the same values, indicating that the particles undergo a similar deformation in all cases. Equivalent diameter and perimeter reduce as impact velocity increases. The particles deform more as particle velocity and temperature increase, resulting in more element deletion. As particles impact and adhere to the substrate, the surface becomes uneven. Despite impacting upon a roughened substrate, an orientation angle near zero degrees suggests a vertical particle deformation. As gas carrier temperature and particle velocity increase and particles experience
more deformation, element deletion due to material damage results in rougher particle edges. Dispersion, which indicates particle edge smoothness, increases as particle velocity and temperature increase as a result of this.

5.4.2 Image Processing of Heated Substrate

Image processing is performed on simulations involving a heated substrate in order to investigate the effect of substrate temperature on particle deformation. Results of the image processing analysis for the simulations involving a heated substrate are given in Figure 41 through Figure 46.
Figure 41: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 581$ m/s, $T_i^{(p)} = 449$ K and $T_s = 453$ K.
Figure 42: Perimeter, Elongation, Dispersion and Roundness histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 581$ m/s, $T_i^{(p)} = 449$ K and $T_s = 453$ K.
Figure 43: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 632 \text{ m/s}$, $T_i^{(p)} = 525 \text{ K}$ and $T_s = 510 \text{ K}$.
Average: 329.1  
Std Dev: 107.2

Average: 1.491  
Std Dev: 0.5985

Average: 0.2030  
Std Dev: 0.2023

Average: 3.063  
Std Dev: 1.523

Figure 44: Perimeter, Elongation, Dispersion and Roundness histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 632$ m/s, $T_i^{(p)} = 525$ K and $T_s = 510$ K.
Figure 45: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 677$ m/s, $T_i^{(p)} = 602$ K and $T_s = 567$ K.
Figure 46: Perimeter, Elongation, Dispersion and Roundness histograms for 100 particle impacts of aluminum with $V_i^{(p)} = 677$ m/s, $T_i^{(p)} = 602$ K and $T_s = 567$ K.

Table 8 lists the averages and standard deviations for all of the parameters in Figure 41 through Figure 46.
<table>
<thead>
<tr>
<th>Sample, Temperature (K)</th>
<th>$T_s = 453 \text{ K}$</th>
<th>$T_s = 510 \text{ K}$</th>
<th>$T_s = 567 \text{ K}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>$V_{i}^{(p)} = 581 \text{ m/s}$</td>
<td>$V_{i}^{(p)} = 632 \text{ m/s}$</td>
<td>$V_{i}^{(p)} = 677 \text{ m/s}$</td>
<td></td>
</tr>
<tr>
<td>$T_i^{(p)} = 449 \text{ K}$</td>
<td>$T_i^{(p)} = 525 \text{ K}$</td>
<td>$T_i^{(p)} = 602 \text{ K}$</td>
<td></td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>FEA Characteristic</th>
<th>Average</th>
<th>Std Dev</th>
<th>Average</th>
<th>Std Dev</th>
<th>Average</th>
<th>Std Dev</th>
</tr>
</thead>
<tbody>
<tr>
<td>Axis Ratio</td>
<td>2.91</td>
<td>1.31</td>
<td>3.09</td>
<td>1.66</td>
<td>3.05</td>
<td>1.50</td>
</tr>
<tr>
<td>Eccentricity</td>
<td>0.90</td>
<td>0.07</td>
<td>0.91</td>
<td>0.08</td>
<td>0.90</td>
<td>0.09</td>
</tr>
<tr>
<td>Equivalent Diameter (pixels)</td>
<td>72.25</td>
<td>22.91</td>
<td>63.37</td>
<td>20.76</td>
<td>58.62</td>
<td>22.08</td>
</tr>
<tr>
<td>Equivalent Diameter (µm)</td>
<td>19.58</td>
<td>6.21</td>
<td>17.17</td>
<td>5.63</td>
<td>15.89</td>
<td>5.98</td>
</tr>
<tr>
<td>Orientation Angle (degrees)</td>
<td>-3.58</td>
<td>21.56</td>
<td>-1.63</td>
<td>19.02</td>
<td>0.99</td>
<td>23.74</td>
</tr>
<tr>
<td>Perimeter (pixels)</td>
<td>344.28</td>
<td>102.10</td>
<td>329.07</td>
<td>107.22</td>
<td>313.28</td>
<td>129.94</td>
</tr>
<tr>
<td>Perimeter (µm)</td>
<td>93.30</td>
<td>27.67</td>
<td>89.18</td>
<td>29.06</td>
<td>84.90</td>
<td>35.22</td>
</tr>
<tr>
<td>Elongation</td>
<td>1.43</td>
<td>0.56</td>
<td>1.49</td>
<td>0.60</td>
<td>1.47</td>
<td>0.61</td>
</tr>
<tr>
<td>Dispersion</td>
<td>0.14</td>
<td>0.13</td>
<td>0.20</td>
<td>0.20</td>
<td>0.31</td>
<td>0.28</td>
</tr>
<tr>
<td>Roundness</td>
<td>2.52</td>
<td>1.06</td>
<td>3.06</td>
<td>1.52</td>
<td>3.21</td>
<td>1.85</td>
</tr>
</tbody>
</table>

Table 8: Average and standard deviations of image processing parameters for each numerical simulation analyzed in Figure 41 through Figure 46.

As with the room temperature substrates, the equivalent diameter and perimeter decrease with increasing gas inlet temperature. The eccentricity and orientation angle are nearly the same for each simulation. The axis ratio and elongation in the $V_{i}^{(p)} = 581 \text{ m/s}$ and $T_i^{(p)} = 449 \text{ K}$ ($T_G = 573 \text{ K}$) case are much closer to the other two cases for elevated substrate temperatures. This data will be compared to data for images of experimental results.

### 5.5 Image Processing of Experimentally Observed Coatings

The image processing procedure described above is applied to SEM images of the actual coatings. SEM’s are taken of aluminum coatings with the respective input parameters in Table 6 and shown in Figure 47.
Figure 47: SEM of aluminum particles coated upon an aluminum substrate with a) $T_G = 573$ K  
b) $T_G = 673$ K  
c) $T_G = 773$ K.

Programs intended to identify boundaries exist but proved difficult to use on these images. In this work, the particle boundaries were identified and drawn manually. Upon identifying the boundaries, the images were transformed to black and white. Particles below a certain size threshold were removed from the images as well in order to prevent small particle edges from distorting the data. Figure 48 shows the black and white images for each experimental condition with and without the partial particles removed.
Figure 48: Black and white experimental images without (left) and with (right) small particles removed.
Each refined image is analyzed for the desired characteristics. The image corresponding to input conditions of $T_G = 573$ K had a magnification of 250× while the images corresponding to $T_G = 673$ K and $T_G = 773$ K had a magnification of 200×. To account for this difference, the perimeter and equivalent diameter, which are based upon pixels, are multiplied by a 1.25 scaling factor for the $T_G = 673$ K and 773 K cases. Figure 49 through Figure 54 show the histograms for the data acquired.

![Histograms](image)

**Figure 49:** Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for experimental conditions of $T_G = 573$ K.

- **a)** Average: 1.832
  - Std Dev: 0.6449

- **b)** Average: 0.7674
  - Std Dev: 0.1503

- **c)** Average: 90.78
  - Std Dev: 25.77

- **d)** Average: 1.050
  - Std Dev: 36.28
Figure 50: Perimeter, Elongation, Dispersion and Roundness histograms for experimental conditions of $T_G = 573$ K.
Figure 51: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for experimental conditions of $T_G = 673$ K.
Figure 52: Perimeter, Elongation, Dispersion and Roundness histograms for experimental conditions of $T_G = 673$ K.

- **Perimeter (a)**: Average: 420.1, Std Dev: 136.2
- **Elongation (b)**: Average: 0.9661, Std Dev: 0.5073
- **Dispersion (c)**: Average: 0.1290, Std Dev: 0.1173
- **Roundness (d)**: Average: 1.893, Std Dev: 0.5955
Figure 53: Axis ratio, Eccentricity, Equivalent Diameter, and Orientation Angle histograms for experimental conditions of $T_G = 773$ K.
Figure 54: Perimeter, Elongation, Dispersion and Roundness histograms for experimental conditions of $T_G = 773$ K.

Table 9 describes the averages and standard deviations of the parameters analyzed in Figure 49 through Figure 54. The 100 µm scale bar associated with Figure 47a spans 325 pixels, giving a conversion of 3.25 pixels/µm. This conversion factor will be used for calculating the equivalent diameter and perimeter in µm.
Table 9: Average and standard deviations of image processing parameters for experimental results analyzed in Figure 49 through Figure 54.

Though almost all of the parameters for the $T_G = 573$ K case are slightly lower than the other two cases, they are well within one standard deviation. The dispersion and orientation angle are nearly the same for all cases. Despite having different impact conditions, the particles show largely the same morphology in all experimental images.

5.6 Comparison of Image Processing Results

Results of all analyzed images and simulations are combined for each quantitative parameter in Figure 55. The error bars indicate one positive or negative standard deviation.
Figure 55: Averages of each quantitative parameter for all simulation and experimental results. Error bars are +/- one standard deviation.
When comparing FEA results to corresponding SEM results, the FEA particles undergo more deformation than what is seen in experiments. Each experimental result has roughly the same values for every parameter. The axis ratio and elongation in the numerical simulations are consistently higher than those found in the experimental images. Though the dispersion and roundness stay roughly constant in the experimental results, they increase with higher temperatures in the numerical simulations. The perimeter and equivalent diameter data for the FEA cases do not fully compare with the SEM data, only proving that the initial FEA particle size was close to the average particle size in the experimental results. Overall, this data suggests that the material damage model presented may require some tweaking, especially at elevated temperatures.
CHAPTER 6. CONCLUSION

High strain rates and temperatures involved in the cold spray process produce an environment where material properties such as flow stress and elastic modulus are sensitive to input gas parameters. In order to properly analyze particle impacts upon a substrate, the momentum and heat transfer between the input gas and particle/substrate material was calculated. Particle velocities and temperatures were determined through a 1D gas-particle flow model in MATLAB. Initial substrate temperature was calculated using a 2D heat transfer analysis between the carrier gas and the substrate. Heat dissipation into the substrate was accounted for by two assumptions: immediate secondary impact of particles or secondary impact after the initial particle has cooled to the substrate temperature.

In this work, a series of single particle impacts followed by two simultaneous impacts was carried out for various conditions related to common cold spray procedures. This model was expanded to simulate 100 successive particle impacts. Computational results were then compared to scanning electron micrographs of aluminum and copper coatings created through the cold spray process. In particular, particle morphology was compared for results with similar input parameters.

FEA results of 3 particle impacts show that as inlet gas temperature \( T_G \) increases, particle deformation and material damage (level of element deletion) increases. This trend is a result of the higher kinetic energies present in the higher gas temperature cases due to higher particle impact velocities \( V_{i(p)} \). As the inter-particle spacing \( \delta \) increases, the initial particle deformation decreases, indicating that deformation and damage are related to the amount of
energy transferred to the particle. In addition, copper particles experience significantly larger deformations than aluminum due to higher densities and temperatures at impact. However, while both aluminum and copper substrates are tested for each particle material, these simulations show that substrate material minimally affects the deformation of the particles when compared to the particle material itself. The softer aluminum substrates result in larger crater depth than those created by impacts upon a copper substrate.

Impacts were simulated upon room temperature substrates as well as substrates that were heated by the incoming gas. Impacts upon a heated substrate produced a higher crater depth than those impacted upon a cold substrate.

In general, the initial particles in the simulations that were allowed to cool before subsequent impacts deformed less than particles that were subsequently impacted immediately after initial deposition. As gas inlet temperature increases, this difference becomes more pronounced. Allowing the initial particle to cool has a large effect on the particle morphology for impacts upon a cold substrate but a slightly smaller effect on impacts upon a hot substrate. The already hot substrate is relatively close in temperature to the initial particle temperature, giving rise to a small temperature change over time. When comparing simulation results to comparative SEM images, simulations involving aluminum impacts compare very well while copper simulations tend to show larger deformations. Aluminum particle simulations show flat particle-particle interfaces that are found in SEM images of deposited aluminum. Copper particle simulations under certain conditions show a conical shape of the initial particle, which is also seen in SEM images of cold sprayed copper. This difference is exaggerated at higher temperatures. The
largely flattened copper particles resemble the extreme deformation found in cold spray experiments as well.

Image processing is utilized to compare experimental coatings with simulations involving 100 successive impacts of aluminum particles upon both room temperature and heated substrates. Increased particle temperature and velocity resulted in more particle deformation and element deletion. SEM images of experimental results showed similar particle morphology regardless of inlet gas temperature $T_G$.

Overall, these multiple particle impacts closely represent particle morphologies that are commonly found in SEM images of cold sprayed metallic coatings. Furthermore, the addition of an intermediate cooling step and the variation of substrate temperature provide a significant change in particle morphology, particularly at elevated temperatures where deformation is greatest. Allowing the initial particle time to cool is essential when simulating particles impacting upon a cold substrate. For heated substrates, this cooling effect has less of an effect. However, as temperature increases, the effect becomes more pronounced. Both the substrate temperature and the condition of the previously impacted particles are crucial when performing simulations with multiple particle impacts.
CHAPTER 7. FUTURE WORK

In this thesis, the deformation of metal particles undergoing high strain rates is studied as a result of varying temperature and velocity. The effect of substrate temperature and application of an intermediate cooling step upon the initial particle have been studied in order to assess the influence of substrate temperature upon the incoming particle deformation. A series of single particle impacts followed by two successive particle impacts have been performed using the finite element method. This three particle model was then extended to deposition of 100 successive particles. Image processing was performed to quantify the relationships between numerically simulated impacts and experimental depositions. The current research can be extended in a number of ways as highlighted below.

1. Due to particle impacting and cooling rates, two assumptions were made: secondary impacts occur either immediately after initial impact or after the initial particle has cooled to the substrate temperature. Particle deformation was compared between these two assumptions and it was found that cooled particles exhibited less deformation than those that were immediately impacted again. Because of this difference, allowing particles to cool before subsequent impacts in the 100 particle simulations may result in particle morphologies that better resemble those found in experimental results.

2. In the copper three particle impacts, the initial particle experiences severe deformation and material damage. Because of this damage, copper proved to be a difficult material to implement into the 100 particle simulation presented in this work. With allowing each particle to
cool before further impacts occur, it may be feasible to model the impacts of copper particles without observing severe material damage.

3. While the time between particle impacts is calculated and two separate assumptions are made (immediate successive impacts and independent impacts), the effect of temperature on multiple impacts is not well known. Development of a systematic analysis would help illuminate the influence of particle temperature and cooling rate upon the deposition of subsequent particles.

4. Originally, the material model produced by Johnson and Cook was created for material undergoing strain rates in the range of $10^3$ 1/s. In cold spray impacts, particles undergo strain rates around $10^6$ 1/s. Developing a material model that extends to the strain rate range occupied by cold spray would help refine the current finite element model.

5. Only copper and aluminum were used as materials for these particle impacts. Other metals such as molybdenum, titanium, and zinc exhibit properties that would be beneficial when applied to the cold spray process. Extension of the multiple particle impact model to different materials could result in a better understanding of material mechanics and determine essential parameters such as critical velocity and input gas temperature.
REFERENCES


