Film Surface Characterization in Cold Spray using Advanced Numerical Modeling and Simulation Techniques

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***Abstract*—In cold spray, even when the initial properties are within the critical values for deposition, the multi-impact process is much more complex to ensure a 100% deposition. In inelastic impact, part of the initial kinetic energy of the particles at impact is lost through plastic deformation. After impact and subsequent restitution, unbounded particles will be ejected through rebound forces or material jetting at erosion. Observation of the final kinetic energies achieved in the particles after impact was used as criteria for a rough estimate of the coating process. The surface roughness increased with reduced deposition efficiency. The interface roughness was less affected by this process. Qualitative comparison to experimental results shows some closer correlations with some of the surfaces of experimentally obtained surfaces in cold spray. This could provide some of the answers to the underlying mechanisms in which the cold spray surfaces are generated.**

Keywords-Multiple particle impact; cold spray; surface roughness; rebound and adhesion energy; deposition efficiency

I. INTRODUCTION

The current desire to have cold spray of functional thin films requires the understanding of the coating process in much finer level. This has the advantage that specific properties can be enhanced in the coatings whilst undesired properties can be minimized or eliminated. In this study we looked at one possibility in which the surface roughness properties of the cold sprayed films are generated. We conducted numerical modeling analysis and further examined experimental data and literature related to the subject.

One of the major challenges in numerical modeling of the cold spray process is that adhesion and cohesion forces are not numerically generated as a result of elements achieving the bonding properties during impact. The user has to design user algorithms in the numerical code to model this process. This could be a tedious task and especially if the major project is not on the implementation of mathematical constitutive equations in the numerical code. This could be regarded as a diversion to the task at hand. Due to these difficulties, one devise a less time consuming methodology which may not be very accurate but can get a very close result and be able to understand the details of the process. It is in this direction that this study used a much simpler method based on the rebound and adhesion energy from kinetic theory.

In this study we examine the possible influence of deposition efficiency variation in the coating process. In order to setup the numerical model, the properties of the particle motion after impact are used as parameters to model their effects on the surface structure.

II. DEPOSITION EFFICIENCY AND CRITICAL VELOCITY

Parameters that have greater impact on surface properties of the coating would be the deposition efficiency and the flux density distribution. A study of rebound energy at various impact velocities within the critical velocity range could give a guide on bonding properties. There is not much understood on the influence of the particle flux on the structure of the coatings generated for the different velocity settings of the transport system, in particular the effect on the surface roughness of the coatings.

At this point we seek to answer the following questions regarding the cold spray coating process: (1) how to get a uniform thickness and smooth surface from cold spray, (2) how to get a rough and porous surface; (3) how to get a dense coating but highly rough surface. We believe different types of coatings are possible with the cold spray method. A detailed understanding of parameters involved for specific type of coating will enable specialized coatings for specific applications. In addition specialized equipment using cold spray method can be designed and further automated for specialized functions.

III. KINETIC THEORY BASED ON CLASSICAL PARTICLES DURING IMPACT

In kinetic theory on cold spray, the rebound energy (E_R) is given as:

$$E_R = \frac{1}{2} e_r m_p V_p^2 \tag{1}$$

where, m_p, ρ_p and V_p are the mass, the density of the particle material and the velocity of the particle at impact respectively. The rebound coefficient e_r for spherical particles initially developed in [1] can be given according to [2] as:

$$e_r = 11.47 \left(\frac{\bar{\sigma}_Y}{E}\right) \left(\frac{\rho_p V_p^2}{\bar{\sigma}_Y}\right)^{\frac{1}{4}} \tag{2}$$

here $\bar{\sigma}_Y$ is the effective yield stress during impact, E is the conversional elastic modulus of the particle and substrate materials. Supersonic particle impact involves shockwave effects and heating which can complicate the impact process. A strain-hardening, strain-rate sensitive, thermal-softening, and deformation localization constitutive model must be considered for the calculation of the effective yield stress $\bar{\sigma}_Y$, which can be given according to the Johnson-Cook plasticity model. The equivalent elastic modulus E can be given as:

$$\frac{1}{E} = \frac{1 \quad \nu_p^2}{E_p} + \frac{1 \quad \nu_s^2}{E_s} \qquad (3)$$

where E_p and ν_p are the elastic modulus and poison's ratio of the particle respectively, E_s and ν_s are the elastic modulus and poison's ratio of the substrate respectively.

In order for the particles to remain bonded after impact, adhesive forces are required. The adhesion energy (E_A) is defined as the energy for detaching the bonded particle from the substrate [2] and is expressed as: $E_A = \alpha E_o$, where E_o is the maximum adhesion energy of a given particle to the substrate; α is the fraction of bonded atoms per unit adhesive interface, and is also called the relative strength of the bond between the particle and substrate (In multiple particles this should be between contacting particles). A relation for the fraction on the bonded atoms was initially developed by Shorshorov and Kharlamov [3]. This relation is later improved by Kurochkin, et al. [1] for the dynamic impact spray process which is based on the solution of the kinetic equation of the interaction of atoms at the phase boundary and is given as:

$$\boldsymbol{\alpha} = \frac{E_A}{E_o} = 1 \quad exp\left\{ \quad \vartheta t_c exp\left[\frac{E_a}{kT_c + (1 \quad e_r) m_a V_p^2/2}\right]\right\} \qquad (4)$$

where ϑ is the natural frequency of eigen-oscillations of the atoms in the crystal lattice, t_c is the contact time, E_a is the activation energy of the chemical bonds, T_c is the contact temperature, k is the Boltzmann constant, e_r is the coefficient of restitution during elastic recovery as given by eqn. (2), m_a is the atomic mass of impacting particle, and V_p is the velocity of impact. The fraction of bonded atoms α is mainly affected by the contact temperature T_c and impact velocity V_p [2]. The given relations indicate that for various materials and particle temperatures there is a critical velocity above which the adhesive strength begins to increase sharply [1].

Previous studies indicate that the maximum adhesion energy can be given as: $E_0 = S_c N_a E_1$, where S_c is the contact area of a single particle to the substrate (or to another particle); N_a is the total number of atoms in the unit contact plane.

According to this model, it is necessary to determine the contact time during which particle plastic flow occurs. Contact time t_c is formally described as the time during which the impact particle decelerates from V_p to zero. In another study an empirical mathematical relation which approximates contact time is given as [4]:

$$t_c = \frac{2\varepsilon_p d_p}{V_p} \qquad (5)$$

where, $\varepsilon_p \quad exp\left(\quad K\frac{H_p}{\rho_p V_p^2}\right)$,$H_p = 6x10^7 Pa$ and $K = 1.4$.

In this kinetic model, the eqn. (2) comprise a term $\frac{\rho_p V_p^2}{\bar{\sigma}_Y}$ which has been termed severity of impact [5]. Therefore the severity of impact is related to the adhesion and rebound. This could be a possibility of this parameter to indicate something about the bonding upon impact. This relation is very simple because many parameters are not involved, such as substrate material properties, impact surface conditions, and so on. The coefficient of restitution has recently been applied to analyze bonding in [16]. There are different methods of quantifying the coefficient of restitution. The coefficient of restitution can also be written as:

$$e_c = \frac{impulse\ during\ restoration}{impulse\ during\ deformation} = \frac{speed\ of\ seperation}{speed\ of\ approach}. \qquad (5)$$

The coefficient in eq. (2) and that in eq. (5) are related as ${e_c}^2 = e_r$. If speed of the substrate at separation is neglected in the criterion, then all particles experiencing $V_{pf} \geq \alpha V_p$ are considered not to have bonded. And since the cutoff criterion covers a range, then the velocity of separation can still be accounted for in the selection criterion. This reduces to:

$$e_c \geq \alpha. \qquad (6)$$

In addition, based on the empirical equations for determining the particle velocity, for a given configuration two parameters were chosen for this study: *critical velocity of impact and critical temperature*. These two parameters are also found in eqn. (4). According to [6, 7] the critical velocity of copper impact on copper particle on a study done using Eulerian model for a 20µm particle in a non adiabatic analysis is said to be 300m/s and for adiabatic 290m/s, while in [8, 9] according to the experimental data for copper impact on copper substrate the critical velocity is said to be 570m/s and critical velocity for copper impact on aluminum is 507m/s. It may seem the 570m/s velocity should be the critical velocity that would account for the particle-particle cohesion, while the 507m/s should be critical velocity for initial particle adhesion to the substrate.

IV. Numerical Simulations

Numerical simulations are a vital tool in modern advanced research and systems design as they are used to predict properties some of which can be difficult to examine experimentally. In addition numerical methods are a quick guide in decision making especially in new equipment design, process optimization or theoretical validation. For example in design costs can be minimized substantially by examining the functional capabilities and existing or available options using numerical methods prior to actual building of experimental models.

The JENano Research Group [10] at the University of Johannesburg is working towards understanding the cold spray for coating of nano-particles to generate thin films and bulk nano-composites and nanostructures. Specifically in this study numerical methods are being applied to investigate various modalities so as to find what parameters can be improved on the coating experimental models. This next section discusses the material constitutive models that were used in the numerical investigations done in the study of film coating process to examine the effects of deposition efficiency in the physical structure.

A. Constitutive Models

In metal plasticity Johnson cook model have been extensively used to define the material behavior. The material model for both the particles and the substrate were described according to Johnson and Cook plasticity model. The stresses are defined according to Von Misses plasticity model.

According to Johnson and Cook the yield flow stress, $\bar{\sigma}$ of the material is define by the expression:

$$\bar{\sigma} = [A + B(\bar{\varepsilon}^{pl})^n]\left[1 + C\, ln\left(\frac{\dot{\bar{\varepsilon}}^{pl}}{\dot{\varepsilon}_0}\right)\right](1 \quad \hat{\theta}^m) \qquad (7)$$

where A, B, n, C, and m are material-related constants, $\bar{\varepsilon}_p$ is the effective plastic strain, $\hat{\theta}$ is the non-dimensional temperature given as:

$$\hat{\theta} \equiv \begin{cases} 0 & for\ T < T_{transition} \\ \frac{(T - T_{transition})}{(T_{melt} - T_{transition})} & for\ T_{transition} \le T \le T_{melt} \\ 1 & for\ T > T_{melt} \end{cases} \qquad (8)$$

Where T is the current temperature, T_{melt} is the melting temperature, and $T_{transition}$ is the transition temperature defined as the one at or below which there is no temperature dependence on the resultant yield stress.

The actual parameters used for the two materials are given in Table I.

TABLE I. MATERIAL PROPERTIES

Material Parameter	Material	
	Copper [11]	*Aluminum [12]*
Density, (Kg/m^3)	8960	2710
Thermal Conductivity, (W/(m°C))	386	220
Specific Heat Capacity, (J/Kg°C)	383	920
Melting Point, (℃)	1083	643
Elastic Modulus, (GPa)	124	65.762
Poisson's ratio	0.34	0.3
JC Plasticity: *A*, (MPa), *B*, (MPa), *n, C, m*	90, 292, 0.31, 0.025, 1.09	148.361, 345.513, 0.183, 0.001, 0.859
JC Damage: d1, d2, d3, d4, d5	0.54, 4.89, -3.03, 0.014, 1.12	0.071, 1.248, -1.142, 0.147, 1
Reference Temperature, ℃	25	25
Reference Strain,($1/s$)	1	1

The coupling in the solving of the heat transfer equations was set such that viscoplastic work is converted into heat according to

$$q = \frac{\beta}{\Delta t}\int_{t}^{t+\Delta t} d\sigma_y\, d\bar{\varepsilon}^{pl} \qquad (9)$$

where $\beta = 0.9$ is the Taylor Quinney constant (inelastic heat constant). Frictional contact was modeled as coulomb friction with 0.2 frictional coefficients.

B. Multi - Particle Simulation

The problem setup was studied using ABAQUS CAE finite elements modeling. This was to study the behavior of the multiple particle impact which is prevalent in generation of coatings using cold gas dynamic spray technology. Initially particles were arranged in a configuration as Figure 1. Particle locations were made to be random by arbitrary positioning. An infinity boundary support on bottom of substrate and on the sides was used. Initial particle velocities and initial temperatures were specified for the problem investigation. Surface interaction was defined using generic contact algorithm. There were no cohesive or adhesive properties enforced during deformation. In view of that the simulation results are only observed in the time of impact and duration of deformation. This is up to the onset of subsequent restitution.

The ABAQUS solver used is the coupled dynamic temp-displacement analysis which accounts for various energy forms in the computation.

After the simulation it was decided to track some elements/nodes which exhibited a different phenomenon from most of the elements to investigate the observed behavior. The elements with increased velocity of restitution were targets for tracking because most elements did not show this behavior. The status was observed at 250ns and again at 300ns for enforcement of the bonding criteria manually.

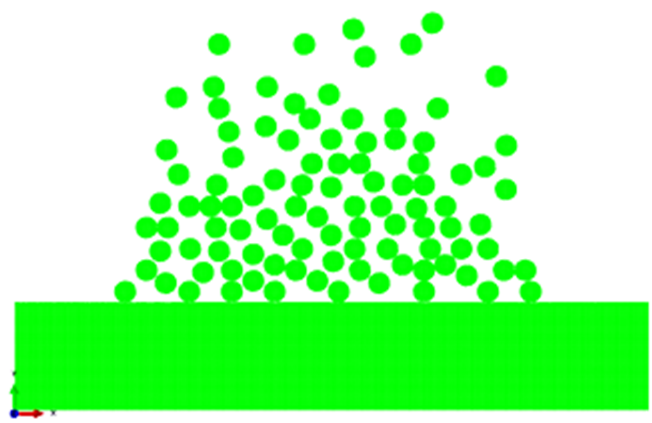


Figure 1. Computational domain, (meshing resolution of 1/20dp, where dp= 10um particle diameter)

V. SIMULATION RESULTS AND SURFACE MODELING CONCEPTS

Figure 2 shows the magnitudes and distribution of the velocity vectors in the x-axis for the depositing particles at 300ns after initial impact. It is easy to see that the particles on the edges of the coating have higher magnitudes with an outward direction components from the impact zone. The surface also shows a somewhat significant velocity vector than that in the interior of the coating.

Figure 3 image shows the magnitudes and distribution of the velocity vectors in the y-axis for the depositing particles at 300ns after initial impact. For these velocity vector components, the particles on the edges of the coating have higher magnitudes with outward direction components from the impact zone. The simulation also shows somewhat significant velocity vector magnitudes on a few surface particles than that in the interior of the coating.

The resultant velocity vectors are shown in the image in Figure 4. The selection procedure used here is based on the final kinetic energy of the particles. Notice that a threshold of 50m/s velocity magnitude was used as a cutoff for selection as either adherence or detached particle. The detached particle was fully removed from the resulting coating. Using such a criterion it is seen that the resultant morphology is highly rough on the surface more than it is on the interface morphology.

The Figure 5 was used in the cutoff criteria. The figure shows the particle status at 250ns after first impact of particles on the substrate. In terms of the velocities upon subsequent restitution, the particles with prominent velocities above 50m/s ($\alpha \geq 0.1$) were removed and considered to have detached from the coating.

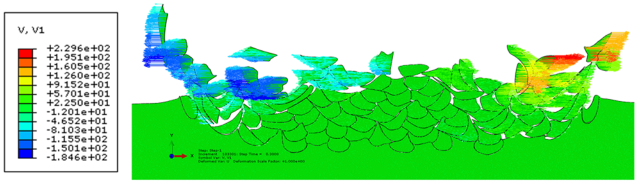


Figure 2. Velocity vectors in the x-direction (m/s)

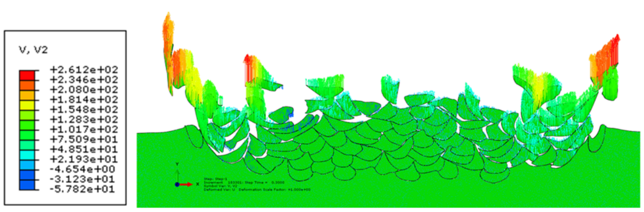


Figure 3. Velocity vectors in the y-direction (m/s)

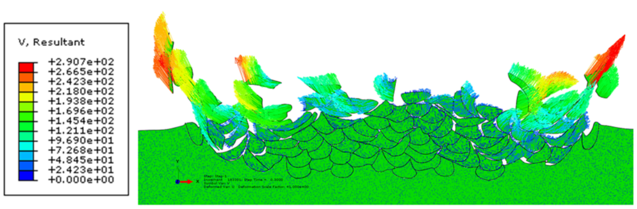


Figure 4. Resultant Velocity Vectors (m/s)

Using this criterion is similar to enforcement of cohesion or bonding at the time of subsequent restitution. The resulting structure is shown in Figure 6.

The Figure 6 shows a highly rough surface as compared to the substrate interface. Porosity is also seen and a region showing highly dense or compact packing of the particles. The deposition efficiency of this numerical modeling approach was at 68%. In addition this problem initially assumed the conditions just before impact were within critical conditions for deposition. This would take care of some un modeled physics in the criterion. The choice of the value of α has not been based on any prior investigation to much with any physical evidence, but was based on the physical observation of the particle status after impact.

VI. Roughness Analysis

"The very first step in surface topography analysis consists of a visualization of the micro geometry, either as single profiles or as surface areas, to provide realistic representations of the surface. The usefulness of such an approach for a qualitative characterization is well recognized: often the image inspection, possibly aided by some enhancement techniques, can be assumed as the only aim of the analysis. Indeed, the image conveys a vast amount of information, which can be easily interpreted by an experienced observer: "even a single profile contains a large amount of relevant information," [13].

The definition of roughness can be found in Smith [13]. Quantitative parameters of the roughness properties and their description are given in Table II according to [13, 14]. Using the image properties given in Table II, the topographic height skewness distribution (Rsk) of the simulated coating comprise of both negative and positive distributions. This results in Kurtosis (Rku) of the topography height distribution being of both spiky and bumpy profile.

The spiky and bumpy surface seem to match experimental evidence obtained with low deposition efficiencies as studied in [15] whose images are shown in Figure 7. Indeed deposition efficiency in both the simulation and experiment changed the topographic distributions and profile in a similar manner.

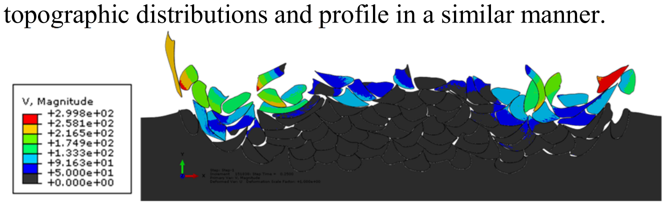


Figure 5. Velocity Magnitude distribution (m/s)

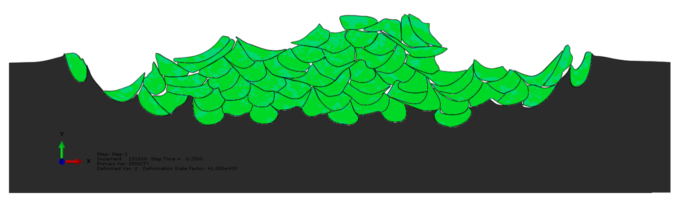
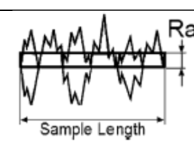
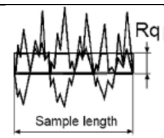
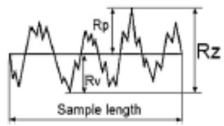
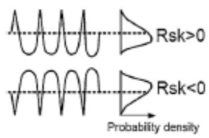
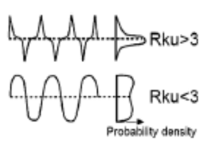


Figure 6. Resultant Coating Structure

TABLE II. Roughness Parameters [13,14]

Symbol	Name (unit) and Definition	illustration
Ra	Arithmetic average roughness (μm) Arithmetic mean of the absolute values of the surfaces departure from the mean	
Rq	Root mean squared (μm)	

	Geometric average value of the profile departure from the mean line within sampling length	
Rz	Maximum peak to valley height (μm) Of the profile within the sampling length	
Rsk	Topography height skewness distribution Measurement of the symmetry of the surface deviation about the mean reference plane. Rsk is negative if the distribution has a longer tail at the lower side of the mean plane and positive if the distribution has a longer tail at the upper side of the mean plane.	
Rku	Kurtosis of the topography height distribution Measurement of the peak or sharpness of the surface height distribution. A spiky surface has a high Rku value and a bumpy surface has a low Rku value.	

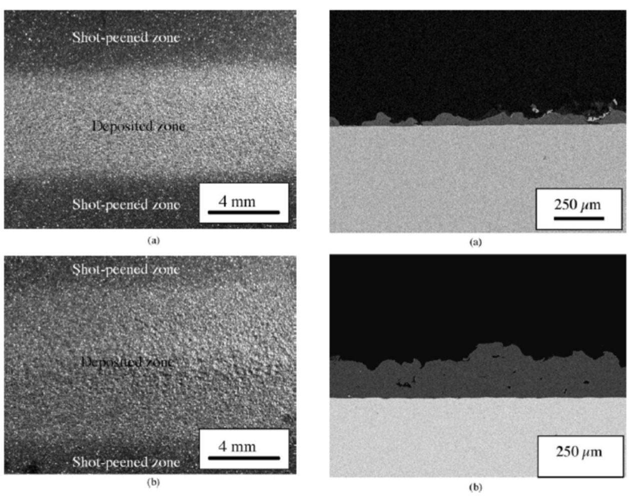


Figure 7. Single pass spraying surfaces (a) 2.9MPa, 300 C; (b) 2.9 MPa, 500 C; Crosssections of spraying traces; (a) multi pass spraying trace at 2.9 MPa, 300 C; (b) single pass spraying trace at 2.9 MPa, 500 C[15].

VII. CONCLUSION

One of the major challenges in numerical modeling of the cold spray is the adhesive and rebound forces which determine particle bonding. In this study the coefficient of restitution was used as a criterion to determine particle bonding in cold spray. A value of 0.1 coefficient of restitution gave a 68% deposition efficiency. It was found that the surface roughness is highly affected by the deposition efficiency. The results also indicates that for the model setup used, the particles at the peripheral are more likely to rebound than those centrally located at impact zone during coating. The results show that the deposition efficiency has a tendency to create artifacts on the surface morphology which drives the roughness parameters of the cold sprayed surfaces. In particular the topographic height skewness, Rsk, and the sharpness of the surface height distributions, Rku, were considered in this qualitative analysis. For the initial conditions given of the materials studied, the bonded particles created a bumpy height profile while the rebound impacts created a spiky height profile. However we believe that other parameters such as the Ra, Rq and Rz parameters (see Table II for definations) are also affected. Future study will look at a quantitative analysis of these roughness properties.

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In 2011, Prof Jen was elected as a Fellow to the American Society of Mechanical Engineers (ASME), which recognized his contributions to the field of thermal science and manufacturing. As stated in the announcement of Prof Jen Fellow status in the 2011 International Mechanical Engineering and Congress Exposition, its states "Tien-Chien Jen has made extensive contributions to the field of mechanical engineering, specifically in the area of machining processes. Examples include, but not limited to, environmentally benign machining, atomic layer deposition, cold gas dynamics spraying, fuel cells and hydrogen technology, batteries, and material processing." Prof Jen has written a total of 198 peer-reviewed articles, including 84 peer-reviewed journal papers, published in many prestigious journals. He also has written six chapters in special topics book.