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Particle Loading Effect in Cold Spray

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Cold gas dynamic spray is a line-of-sight, high-rate material deposition process that uses a supersonic flow to accelerate small particles (micron-sized) above a material-dependent critical velocity. When the particles impact the substrate, they plastically deform and bond to form a coating. The objective of this research is to investigate the influence of the particle mass flow rate on the properties of coatings sprayed using the cold spray process. Varying the mass flow rate at which the feedstock particles are fed into the carrier gas stream can change the thickness of the coating. It was shown that poor coating quality (peeling) was not a result of flow saturation but, instead, the result of excessive particle bombardment per unit area on the substrate. By increasing the travel speed of the substrate, this can be overcome and well-bonded dense coatings can be achieved. It has also been shown that by heating the carrier gas flow poor coating quality is avoided.

Keywords critical impact velocity, deposition efficiency, peeling, powder mass flow rate

1. Introduction

The cold gas dynamic spraying (CGDS) process, also referred to as cold spray, is the latest of the thermal spray processes. Although similar processes were proposed as early as in the 1950s (Ref 1, 2), cold spray has only emerged in the last decade. Early skepticism toward the capability and viability of the CGDS process has slowly faded away over the last few years and has been replaced by worldwide interest in the process. Global efforts have lead to an improved understanding of the process, and its potential has been explored by research teams such as the ones in Sandia National Laboratories (Ref 3) and Delphi Research Laboratories in the United States (Ref 2, 4), the University of Bunderswehr in Germany (Ref 5-7), and the Institute of Theoretical and Applied Mechanics of the Russian Academy of Science in Russia (Ref 8-10), to name a few. Those efforts are helping cold spray to find its niche among the other thermal spray processes such as plasma and high-velocity oxy-fuel spraying.

As opposed to other thermal spray processes, such as plasma and high-velocity oxygen-fuel spraying, cold spray does not require melting or heating/softening of the powder to be sprayed on a substrate (Ref 8, 11). This line-of-sight process accelerates solid particles by a supersonic flow above a critical impact velocity. At impact, the particle kinetic energy is used to plastically deform the particle and disrupt the thin film oxide surfaces. Consequently, it results in intimate conformal contact between the substrate and the particle under a very high localized pressure, forming a coating (Ref 3, 12). However, if the impact velocity is below the critical impact velocity of the material to be sprayed, the particle bounces back from the substrate. Inert gases such as helium or nitrogen are typically used in cold spray to propel the solid particles in the velocity range between 300 and 1000 m/s (Ref 2-4, 6).

Given that there is no major heating of the particles and that inert gases are used as the propellant, cold spray is often referred to as a solid-state process in which high-temperature transformations such as recrystallization and oxidation are minimized. This represents a major advantage over other existing coating techniques from the standpoint of controlling the coating chemical composition and microstructure (Ref 11, 12).

The thick, dense coatings produced by the cold spray process can yield many benefits, and their potential applications vary. It is foreseen that cold spray could be used to apply coatings to large surfaces in many automotive, aeronautics, and aerospace applications. As a result, development in these areas requires ensuring that the coating properties are those needed. However, the ability to achieve the proper coating properties for in-service conditions does not guarantee that the process will be used. For most applications, the overriding consideration is economics. The chosen coating process must be economically viable, and to achieve this the ability to coat large surfaces rapidly must exist. Consequently, the process deposition rate must be maximized.

In this article, a study of the effect of the powder deposition rate, more specifically the particle mass flux, of copper powder on aluminum substrates using the cold spray process was performed. The copper coatings produced, using different powder deposition rates, are characterized using scanning electron microscopy (SEM) to examine microstructure and thickness. To compare the effect of the process gas stagnation temperature, the propellant gas is heated to determine whether increased temperature can improve coating quality and deposition rate. The deposition efficiency is also monitored. The particle impact velocity is modeled using a validated mathematical model (Ref 13) for the different cases to assess the powder mass flux.

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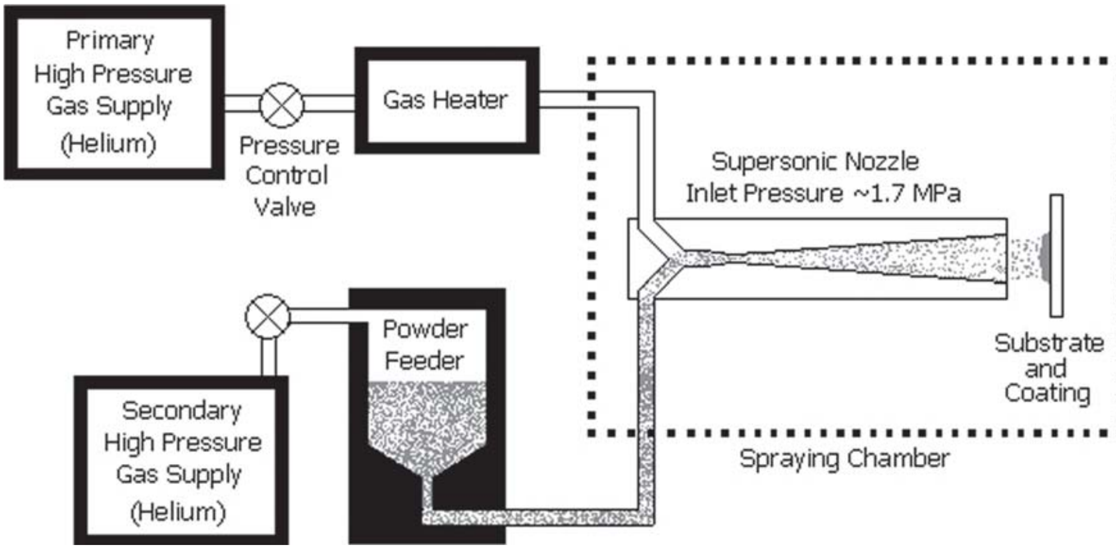


Fig. 1 Schematic of the cold spray facility at the University of Ottawa

2. Experimental Setup

2.1 Spraying System

The computerized spraying system used in this work was developed at the University of Ottawa cold spray facility and has been used extensively to spray a variety of coatings (Ref 12-14). The setup, consisting of a gas control system, a commercial powder feeder (1264 powder feeder; Praxair, Danbury, CT), a spray gun, and an enclosed spraying chamber, is depicted schematically in Fig. 1. The gun has a converging-diverging nozzle that allows the gas to reach the high velocities required in the process. The nozzle has been designed using a validated mathematical model (Ref 15) to eradicate the presence of shock waves inside the nozzle. The system incorporates two probes that measure the temperature and pressure before the converging section. The substrate holder is motorized to allow a variable traverse velocity.

2.2 Operating Parameters

Helium, due to its aerodynamic properties, was used as the propellant gas for all the tests. The stagnation pressure and temperature were kept constant at 1.7 MPa and 5 °C, respectively. Eight different powder mass flow rates, 0.9, 1.5, 1.9, 2.4, 3.5, 4.1, 4.7, and 5 g/min, were used to study their effect and impact on the process performances and single-layer coatings produced with a nozzle lateral traverse speed of 3.5 mm/s. Additional trials were conducted to determine the effect of temperature coating quality and thickness. Coatings were produced through identical procedures with a powder mass flow rate of 4.1 g/min and a carrier gas temperature of 400 °C.

2.3 Sample Preparation, Powder, and Coating Characterization

Prior to the spraying, aluminum substrates were grit-blasted using silica (quartz) grit (20-mesh, 24-grit). A commercially available particle size analyzer (LS; Coulter at UC Davis, CA) was used to determine the particle size distributions of the powders. The morphology of the powders and the microstructures of the coatings were examined using an Hitachi (Tokyo, Japan) S-2250N scanning electron microscope. Prior to SEM observations, coating samples were sectioned from a transverse section and were prepared by standard metallographic techniques. The deposition efficiency was evaluated by weighing the substrate before and after the spraying (P-214; Denver Instruments, Denver, CO). By using the measured powder mass flow rate, the mass accumulated on the substrate, and the duration of spraying, the deposition efficiency was determined.

Adhesion testing was also performed on the coatings according to the ASTM C633-01 standard (Ref 16).

3. Mathematical Model

A validated mathematical model was used to study and predict the particle impact velocity for each case. The assumptions used to model the gas flow fields, the solid particle motion, and the interaction between the two phases, are presented. It was assumed that the flow obeys the continuum, and is compressible and turbulent. The fluid was assumed to be a Newtonian viscous fluid, and to follow the perfect gas law. The Stokes hypothesis was also assumed to be valid. The Morkovin hypothesis was used. Consequently, the Reynolds-average Navier-Stokes equations were used to describe the flow field, and the turbulence models remained valid with the empirical data taken from incompressible flow experiments. Triple turbulent correlations were assumed to be negligible, and the Boussinesq assumption was used. As a result, the $\kappa - \varepsilon$ turbulence model (Ref 17) was used to include and account for the effect of turbulence in the flow field.

For modeling purposes, the nozzle was assumed to be axisymmetric. The injected particles were modeled as perfect spheres, and particle-particle interactions were not considered. The particle-gas interactions were included in the momentum

Table 1 Powder size of copper

Diff. volume, %	10	25	50	75	90
Size, μm	<17.1	<24.6	<39.6	<58.0	<80.1

and energy equation using the particle-in-cell method. It could be foreseen that the exact geometry of the powder particles would not be spherical. However, the drag coefficient on nonspherical particles was larger than that on spherical ones. As a result, the velocity of the particles predicted by the model will establish the lower velocity limit attained by the powder particle.

Based on the preceding assumptions, the continuity, momentum, and energy conservation equations in their Reynolds-average general conservative form (including source terms for the gas-particle interactions) were used, as well as the turbulence kinetic energy and the turbulence energy dissipation rate equations. This system of equations was closed using the perfect gas law. The motion of the solid particles was tracked using Newton's second law of motion in a Eulerian-Lagrangian scheme.

The mathematical model has been validated using an optical diagnostic method. Details of the numerical method and procedure as well as the validation are given elsewhere (Ref 15).

4. Results

4.1 Powder Characterization

Pure copper powders (water atomized, 99.9% pure) were used and injected into the nozzle. The choice of copper was made in view of both the feasibility of the coating formation by cold gas spraying and the availability of the relevant material data in the literature (Ref 18). The particle size distribution obtained is shown in Table 1. It can be seen that the distribution of particle sizes varies from 23 to 56 μm.

The morphology of the atomized powder is shown in Fig. 2. The copper powder used for this study was nonspherical. This result reveals that the mathematical model will only predict the velocity of the slowest (spherical) particles for each particle diameter.

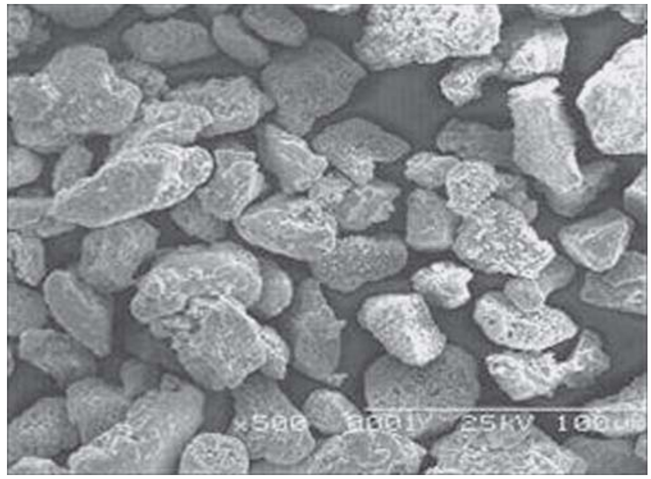


Fig. 2 Powder morphology

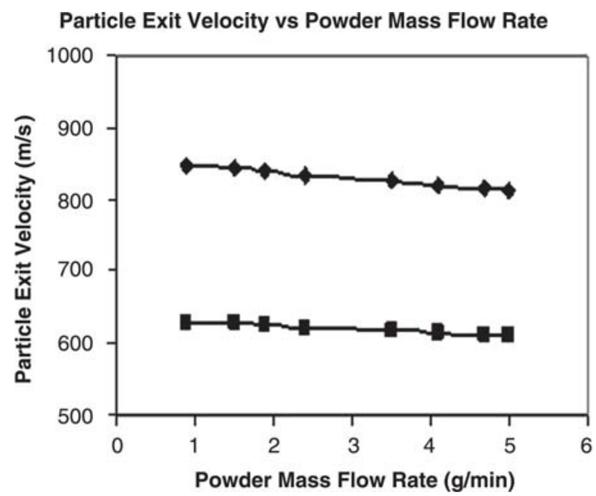


Fig. 3 Modeling results: particle impact velocity for the various powder mass flow rate used. The top line represents the smaller particles, while the bottom line represents the larger particles.

4.2 Modeling Results

During the spraying process, the copper particles must reach the material-dependent critical velocity (440–500 m/s for copper) to gain enough kinetic energy to undergo plastic deformation at impact and form a coating (Ref 8, 19). Furthermore, a shock wave is inevitably formed when the supersonic jet stream emerging from the nozzle comes into contact with the substrate to decelerate the flow to zero velocity at the stagnation point on the substrate (Ref 13). Larger and heavier particles easily maintain their momentum and are less likely than smaller light particles to decelerate across the shock. They will, therefore, maintain enough kinetic energy to impact the substrate to form a coating. The numerical modeling results of the nozzle gas flow and particle acceleration using the maximum and minimum diameters of the particle size distribution used are presented in Fig. 3. It is shown that the larger particles exit the nozzle at a velocity >600 m/s, while the smaller particles exit the nozzle at velocities >800 m/s. The loading effect of the particle at any powder mass flow rate is small according to those results. It is concluded that the vast majority of the particles should be able to reach and maintain a velocity higher than the critical velocity across the shock wave for the entire powder mass flow rate used in this study. It is therefore expected that the process will achieve good deposition efficiency.

4.3 Coating Characterization

It has been established that the coatings produced by cold spray are built according to a variety of particle-particle and particle-substrate interactions, which are similar to those found in explosive compaction (Ref 6, 11). These interactions are divided into distinctive regions as the particles pile up on the substrate (Ref 20). In the first region, close to the substrate, the first particles impact the surface of the substrate, crater it, deform plastically and mechanically, or metallically bond to the substrate under the high local contact pressure. The second region is built

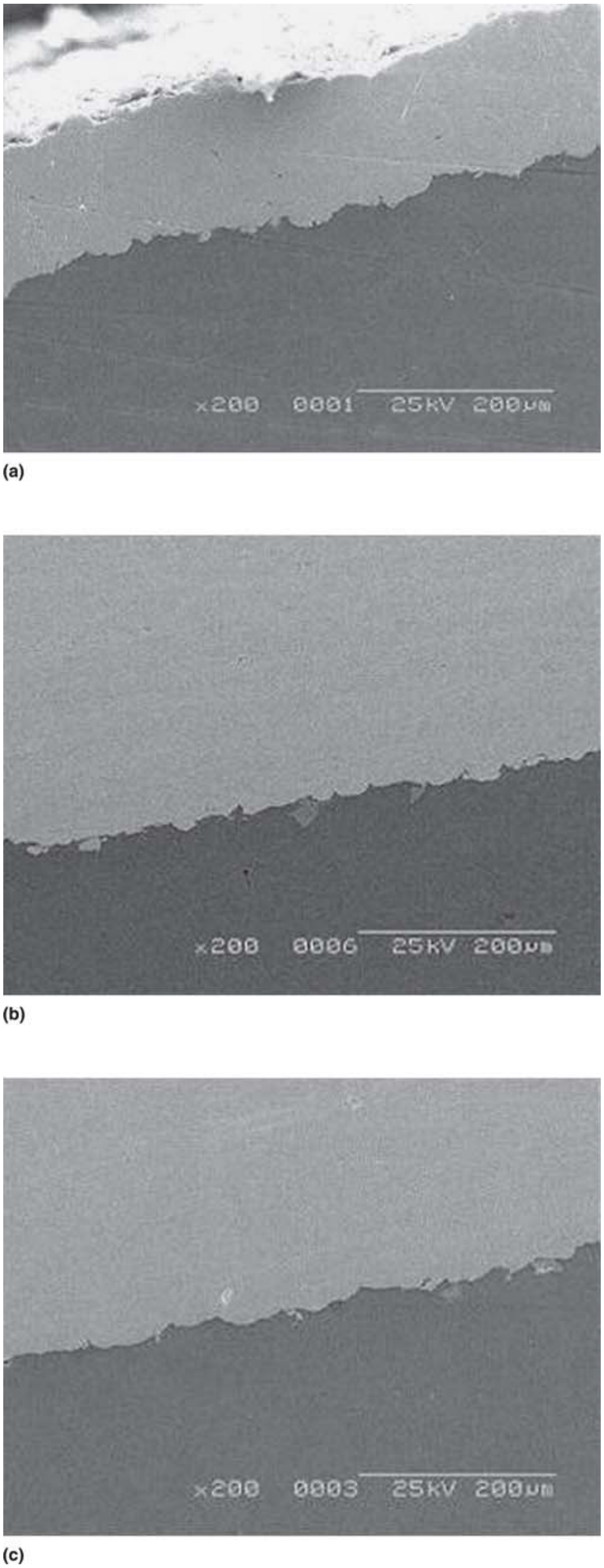


Fig. 4 Cold spray coatings: powder mass flow rate of (a) 0.9 g/min, (b) 3.5 g/min, and (c) 4.2 g/min

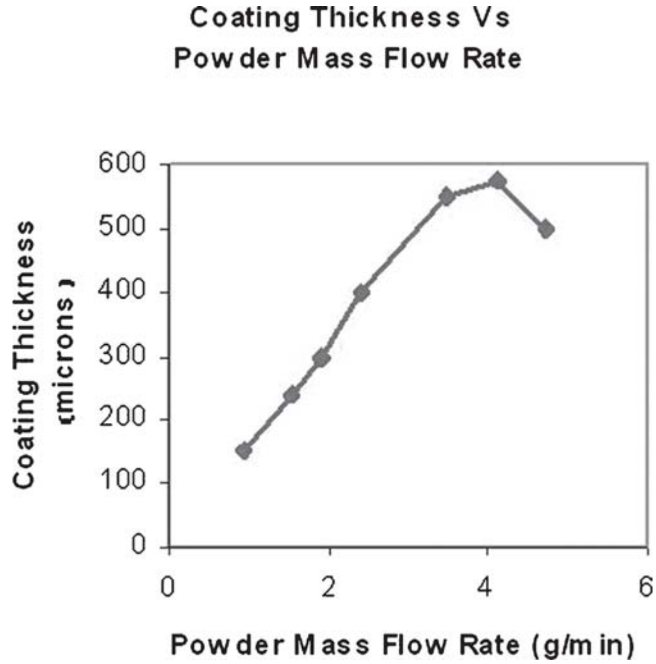


Fig. 5 Cold spray coating thicknesses as a function of the powder mass flow rate and a propellant gas temperature of 5 °C

directly on top of the first one and experiences two different behaviors according to the particle impact velocity. For low impact velocity, minimal particle deformation and little or no bonding occur. The particles realign themselves through rotation and direction to follow the path of least resistance. However, if the kinetic energy of the particle is large enough, the particles experience large deformation, and significant bonding between the particles is found.

Figure 4 shows SEM images of the copper coatings for the powder mass flow rates of 0.9, 3.5, and 4.1 g/min. The images reveal that the coatings are dense with little or no porosity or cracks. This confirms that the particles have been accelerated sufficiently to achieve plastic deformation on impact. Hence the quality of the coating is considered to be good. Also, the substrate-coating interface between the copper coating and the aluminum substrate is easily visible and presents no defect such as pores or cracks.

Figure 5 shows a plot of the coating thickness as determined from the SEM images of the various coatings as a function of the powder mass flow rate used. Coating thicknesses were determined to range from 150 μm (for a powder mass flow rate of 0.9 g/min) to 575 μm (for a powder mass flow rate of 4.1 g/min), increasing with the powder mass flow rate and while the propellant gas was unheated. It is shown that the maximum thickness is reached at 4.1 g/min and that there is then a slight decrease at 4.7 g/min. Above 4.7 g/min, the resulting coatings peeled off the substrate near the coating-substrate interface, as depicted in Fig. 6.

The mass of the copper deposited on the substrate per unit length of the coatings, shown in Fig. 7, indicates findings similar to those of the coating thickness as a function of powder mass flow rate, confirming a constant powder mass flow rate through the spraying process while the propellant gas is unheated. As the

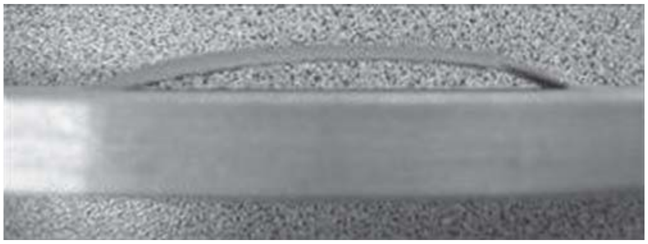


Fig. 6 Cold spray coating sprayed at a powder mass flow rate of 5 g/min and a propellant gas temperature of 5 °C

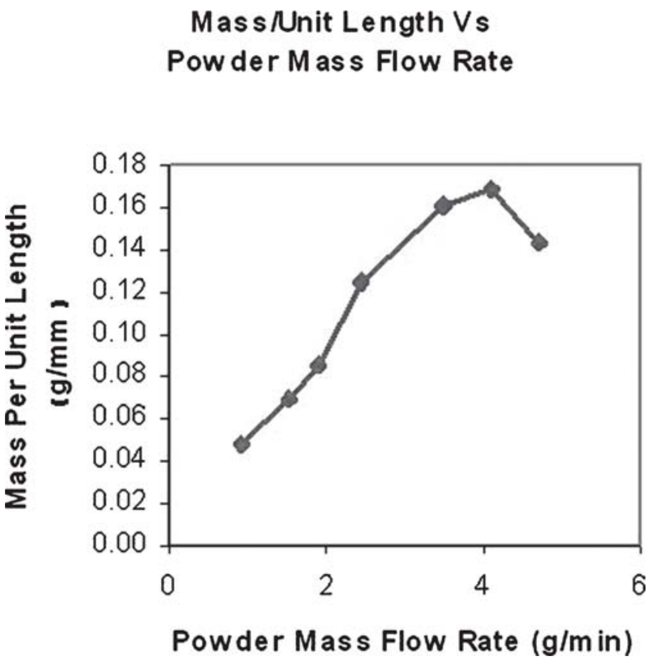


Fig. 7 Cold spray coatings mass per unit length as a function of the powder mass flow rate and a propellant gas temperature of 5 °C

powder mass flow rate was increased, there was an increase in the mass of copper deposited on the substrate until a maximum was reached at a feed rate of 4.1 g/min, at which point the mass deposited decreased as the powder mass flow rate was increased.

The bond strengths obtained from the copper coatings that did not originally peel and were applied with unheated propellant gas ranged from 30 to 35 Mpa, which is in accordance with the ASTM C633-01 standard. This is comparable to the results of other studies in which bond strengths of 30 to 40 MPa have been achieved (Ref 21).

Figure 8 presents the deposition efficiency as a function of the mass flow rate of the powder injected into the nozzle. As Fig. 8 indicates, all coatings resulted in average deposition efficiencies of 85 to 90%.

The peeling of the coating at the higher powder mass flow rate is now examined. Peeling can be said to have occurred after the particles had adhered to the substrate (i.e., shortly after the local spraying process) because a coating layer was observed on the substrate under the peeled portion of the coating. It is unlikely that a coating would originally form in a bow shape such

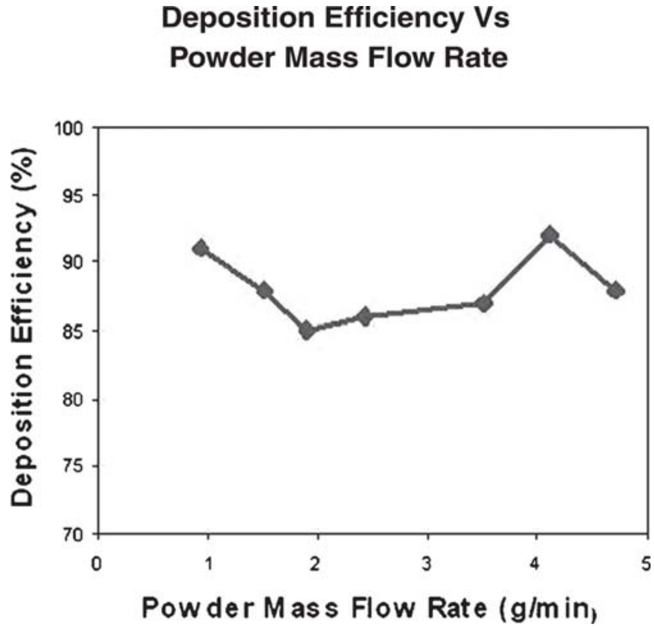


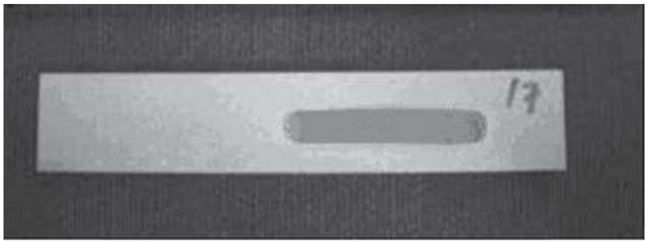
Fig. 8 Cold spray deposition efficiency as a function of the powder mass flow rate and a propellant gas temperature of 5 °C

as the one presented in Fig. 6, because there would be nothing for the particles to initially adhere to. Consequently, it is concluded that the coating peeled progressively as the coating was being formed or after the spraying was completed.

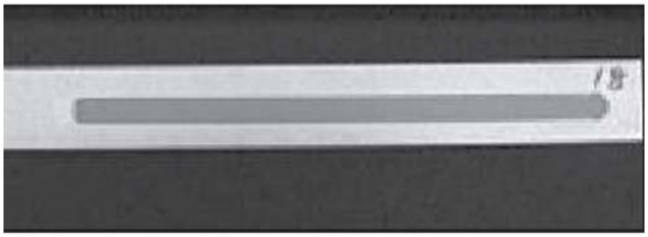
The coating did not peel due to the saturation of the flow. Saturating the carrier gas flow with particles makes the individual particles unable to reach the velocity required to have enough kinetic energy to deform on the substrate and bond properly to form a coating. In the present case, modeling results indicate that at the higher powder mass flow rates the particles are able to maintain a velocity higher than the critical velocity. However, as a result of the higher powder mass flow rate, too many particles impact the substrate over a given surface area per unit of time. This constant bombardment of particles on the same substrate area results in an increase in residual stresses in the coating (i.e., the shot-peening effect). Based on theoretical work (Ref 7), it is also suggested that there is localized melting of the particles on impact. When cooling occurs, the particle will experience localized shrinkage, which would lead to residual stress. It is then apparent that an increase in impacting particles results in greater residual stress in the coating. It is believed that at a threshold these stresses caused the coating to peel near the substrate-coating interface.

The modeling results suggest that the powder mass flow rate does not saturate the gas flow and was verified experimentally. To verify the results, the relative lateral travel speed between the substrate and the nozzle was increased to 6 mm/s using the same powder mass flow rate (5 g/min) as in the cases in which the coating peeled off the substrate. The objective of this is to indicate that reducing the number of particles impacting a given surface area reduces the imposed residual stress and peeling is not observed.

For the case of the increased relative lateral travel speed, a coating that did not peel and that was as dense as the other pre-



(a)



(b)

Fig. 9 Resulting coating that peeled at higher powder mass flow rate (a) and then did not peel at higher substrate speed (b)

viously produced was observed. Referring to Fig. 9(b), one can note that the coating produced at the increased relative substrate-nozzle speed produces a coating that is longer and that does not peel in contrast to Fig. 6 and 9(a). Figure 6 and 9(a) are images of the same coating taken from different angles.

It is noted that the coating that peeled adhered to the substrate at the start of the spray and at the finish. These two instances can be associated with the short transient period resulting from the activation and termination, respectively, of the coating process. Before reaching the steady state, there is a ramping period in which the powder flow rate is lower until it reaches the desired setting and occurs in reverse as the process is shut off. In these instances, a coating that adheres to the substrate is created. The fact that peeling did not occur at those locations confirms that the peeling is caused by extra particle bombardment resulting in residual stresses in the coating.

The trials conducted previously at a powder mass flow rate of 4.1 g/min were repeated with elevated propellant gas temperatures (400 °C). The effect of the increased particle mass flow rate causing the coating to peel was not observed. For all cases, the coatings produced were thick, dense, and fully adhered to the substrate. It was possible to increase the powder mass flow rate in excess of 10 g/min and to create successful coatings at higher propellant gas temperatures. At powder mass flow rates >10 g/min the coating deposition rate was such that the coating thickness became large enough to block the exit of the nozzle. To alleviate this problem, the stand-off distance could have been increased; however, this would then change the spraying parameters, and the correlations with previous trials would not be valid. At higher temperatures, it is believed that there may be a softening of the particles allowing for more particles to adhere to the substrate.

Adhesion testing was performed on the copper trials of unheated propellant gas flows at powder mass flow rates of 3.5 and 4.1 g/min. The bond strength was also tested at an elevated propellant gas temperature (400 °C) and a powder mass flow rate of 4.1 g/min. At these elevated temperatures, increased bond strength was observed. Results of 40 to 53 MPa were obtained, which is a higher range than those acquired from the unheated samples (30–35 MPa). The higher results may be due to superior adhesion resulting from the higher gas temperature. The increased gas temperature may lead to softening of the copper particles and therefore to a greater ease of particle plastic deformation. This could result in a higher quality of coating and could explain the enhanced adhesion.

5. Conclusions

The cold spray process is capable of creating very dense and thick copper coatings on aluminum substrates. Varying the mass flow rate at which the feedstock particles are fed into the carrier gas stream can change the thickness of the coating. Increasing the powder feed rate increases the thickness of the coating linearly until a maximum powder mass flow rate is reached for which there are too many particles impacting the surface of the substrate, resulting in excessive residual stresses causing the coating to peel. It was shown that the peeling was not a result of flow saturation but, instead, of excessive particle bombardment per unit area of the substrate. By increasing the travel speed of the substrate or elevating the propellant gas temperature, this can be overcome, and once again well-bonded dense coatings are achieved. Increasing the propellant gas temperature creates thicker coatings with higher coating adhesion when compared with identical nonheated coatings. It is also possible to increase the powder mass flow rate to create thick coatings when the gas is heated and to not observe the peeling of the coating.

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