Available online at www.sciencedirect.com
ScienceDirect
Surface & Coatings Technology 201 (2006) 1942–1947
SURFACE & COATINGS TECHNOLOGY
www.elsevier.com/locate/surfcoat

Critical particle velocity under cold spray conditions

François Raletz [a,b,*], Michel Vardelle [a], Guillaume Ezo'o [b]

[a] SPCTS UMR-CNRS 6638, Université de Limoges, 123 Avenue Albert Thomas, 87060 Limoges, France
[b] CRITT METALL 2T-LORIUS, PIA des 3 Frontières, 54400, Longwy, France

Available online 6 June 2006

Abstract

Cold spray process is an emerging technique that produces high density coatings. Particles (1 to 50 μm in diameter) are carried by a supersonic gas stream through a de Laval nozzle and, finally, impact on a substrate with high kinetic energy. Low gas temperatures (<600 °C) make it possible to maintain sprayed material in solid state during the whole process. If the particles reach a given velocity, called "critical velocity", they can bind to the surface and create a coating. This velocity is clearly dependent on both sprayed material and substrate properties. This work presents an imaging technique that allows a fast measurement of critical velocity. The measuring method is first evaluated by comparing the critical velocity of copper (sprayed on copper substrate) found in the literature, with the measured one. Its accuracy is then tested with other materials and, finally, some improvements of the method are proposed.

Keywords: Critical velocity measurement; Cold gas spray method; Copper coating; Nickel coating

1. Introduction

Contrary to thermal spray technologies, the cold gas dynamic spraying process, CGDS presented in Fig. 1, makes it possible to manufacture coatings by using very little electric or calorific energy. In this process, the coating formation relies on (i) the kinetic energy of the particles propelled towards the substrate with a velocity ranging between 300 and 1200 m/s; (ii) the ability of particles to deform during the impact process [1]. As the particles undergo no phase change during their flight between the powder feeder and the substrate and, as the gases used in the process are generally inert with a temperature lower than 600 °C, the particles are not subjected to chemical reaction with the gas phase.

Table 1 compares some characteristic parameters of several thermal spray techniques.

The cold spray process is characterized by an impact critical velocity below which no particle adhesion to the substrate is possible. It has been experimentally shown that this velocity depends both on particle and substrate nature and properties. For instance the critical velocity varies when the same material is sprayed on two substrates that have the same chemical composition but have undergone different heat treatments.

Therefore, the knowledge of this velocity is a key point to determine the optimized spraying parameters and reduce the manufacturing cost by increasing the deposition efficiency. The critical velocity can be predicted thanks to mathematical models [2]. However the latter require the knowledge of the properties of

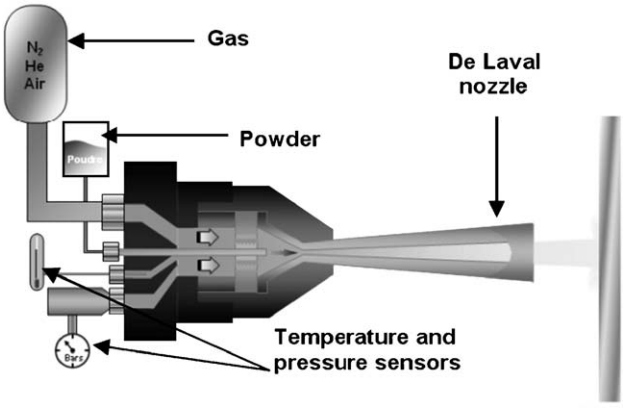

Fig. 1. Cold spray principle.

* Corresponding author. CRITT METALL 2T-LORIUS, PIA des 3 Frontières, 54400, Longwy, France. Tel.: +33 382 23 02 38; fax: +33 382 23 02 42.
E-mail address: francois.raletz@crittlorius.com (F. Raletz).

0257-8972/$ - see front matter © 2006 Elsevier B.V. All rights reserved.
doi:10.1016/j.surfcoat.2006.04.061

Table 1
Characteristic parameters of several thermal spray techniques

	Wire arc	Plasma	HVOF	CGDS
Jet temperature *K*)	~6000	~13,000	~5500	300–900
Jet velocity (m/s)	50–100	800–1500	1000–2000	1000–2500
Gas	Air, N_2, Ar	Ar, H_2, N_2, He	CH_4, C_3H_6, H_2	Air, N_2, He
Gas flow rates (Nlm)	500–3000	40–150	400–1100	1000–3300
Power (kW)	2–5	40–200	150–300	5–10
Feedstock (g/mn)	150–2000	10–80	15–50	20–80
Deposit density (%)	80–95	90–95	>95	>95

materials and are generally limited to pure metals. It can also be determined by using laser Doppler anemometry to measure particle velocity and by associating the measured data with the deposition efficiency of the process [3]. However, this method is rather time-consuming and requires expensive measurement apparatus.

The objective of this study is to present two simple and rather fast methods to determine the critical velocity. The first method uses an imaging technique to determine the distribution of the sprayed particles prior to their impact on the substrate. The second method is based on the calculation of the particle impact velocity from a one-dimensional isentropic model and the measure of the deposition efficiency.

2. Experimental procedure

2.1. Spraying set up

Cold spraying was performed with the Kinetic 3000 M system from CGT GmbH (Wernher-von-Braun-Str. 84539 Ampfing) using a de Laval nozzle (throat diameter: 2.6 mm) and nitrogen as propellant gas. The system controller ensured the reproducibility of the deposition operation and repeatable coatings. The powder feeding rate was sufficiently low (5 to 10 g/min) to have a low loading of the gas phase by particulates and, so, facilitate the velocity measurements on in-flight particles. In cold spraying, the loading effect is less significant than under plasma spray conditions because of the higher gas flow rates (1000–3300 Nlm).

The distance between the nozzle exit of the spray gun and substrate was 30 mm and the gun remained fixed during spraying operation.

When using the imaging technique, the experiments started with low pressure and temperature of propellant gas. The latter was maintained constant during the whole procedure. The combination of a low gas pressure (2 bars) and low gas temperature (150 °C) made it possible to produce a particle spray jet with a velocity below the critical velocity. Then, the gas pressure was gradually increased (2-bars steep), until particles started to stick on the substrate as the impact velocity of the fastest particles was higher than the critical velocity. The formation of the first layer of coating was controlled by the imaging technique.

2.2. Set up for in-flight particle diagnostic

Under CGDS conditions, the main problem for the measurement of particle velocity by an imaging technique is the low temperature of particles. To make up for the lack of brightness, an external illumination is necessary. In this study, the measurements of in-flight particle parameters were done (in Fig. 2) using a SprayWatch equipment (Oseir, Osuusmyllynkatu 13FIN-33700 Tampere Finland) that associates a fast-shutter CCD camera with a high-power pulsed laser diode (HiWatch). The CCD camera sensor had a resolution of 600×480 pixels and the measurement volume was about 20 mm×16 mm×14 mm which was sufficient to cover the whole width of the particle spray jet. During the time of the shutter opening (about 100 μs), the particle spray jet was illuminated by three consecutive laser pulses. The length between the three light points on the CCD detector was measured by an image-processing algorithm and converted into velocity by dividing it by the pulse time-interval. The accuracy of this method that does not require any calibration is about 2% [4,5].

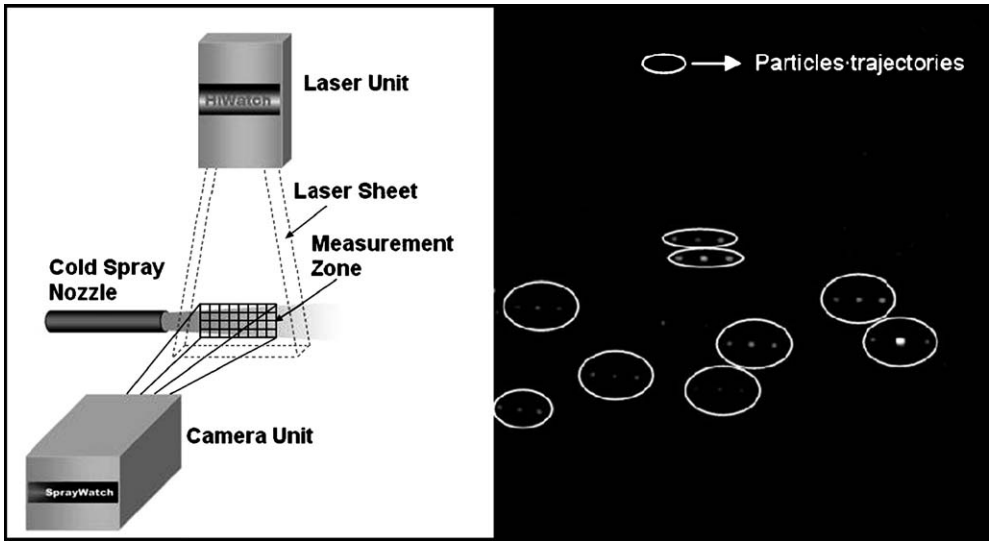

Fig. 2. The laser sheet spray imaging geometry (left picture) and cold spray particles lighted using a laser diode laser (right picture).

The particle velocity measurements were carried out at 16 mm upstream from the substrate and image height was about 20 mm.

3. Determination of the critical velocity by using an imaging technique

3.1. Procedure

As explained (Section 1), the particles start to stick on the surface when their impact velocity is higher than the so-called critical velocity. If particles impinge on the surface with a velocity lower than this critical velocity, they rebound on the surface.

Since the original powder exhibits a particle size range, the particle spray jet exhibits a velocity range and, at low gas pressure, only the finest and therefore lightest particles reach the critical velocity. When the gas velocity is increased, a larger number of particles reach this velocity and the deposition efficiency increases as the number of particles that rebound on the surface decreases.

The first method used to estimate the critical velocity is based on the experimental observation of the particles rebounding the surface by using the SprayWach system that makes it possible to determine particle velocity and flow distribution.

Since the velocity of the rebounding particles is about ten times lower than the velocity of the impacting particles, the system cannot calculate the two types of velocities. With regular laser set-up, time between three laser pulses is much too short to produce three separated points with one bouncing particles and so the calculation algorithm will not consider this signal as a particle. Evaluation of particle flow is done with another method (each single luminous point is considered) and, here, bouncing particles flow is measured.

The presented results are averages made on about fifteen images.

3.2. Example: copper particles sprayed on copper substrate

The pictures of Fig. 3 show the particle spray jet impacting on the substrate for different propellant gas pressures (Table 2).

Table 2
Average particle velocity versus propellant gas pressure

	Propellant gas pressure (10^5 Pa)	Propellant gas temperature (°C)	Particle average velocity (m/s)	Particle velocity Standard deviation (m/s)
A	2	150	271	35
B	5	150	359	41
C	10	150	430	46

The particle size of the original powder ranged between 10 μm and 33 μm. As expected, the particle flow distribution clearly depends on the gas flow velocity.

Three typical cases are illustrated in Fig. 3.

-A The gas pressure is fixed at 2.105 Pa and the velocity of the propellant gas is relatively low. This results in a rather broad particle spray jet (about two times larger than it is in correct CGDS conditions). The impacting particles rebound over the whole substrate surface.

-B The operating conditions correspond to the ones normally used with the spray system of this study. The gas pressure is fixed at 5.105 Pa. The width of the particle spray jet (at 27 mm from the nozzle exit) is about 6 mm and the particles have straight trajectories. However, still some particles have velocities below the criticzal one as shown it can be seen in Fig. 3B. The particles that rebound on the surface are principally located in the fringes of the gas jet.

-C The gas pressure is increased up to 10.105 Pa and the gas velocity is with regard or respect to. The width of the particle spray jet is about the same than in case B but most of the particles stick on the surface after impact.

The radial distributions of the sprayed particles for the three gas pressure conditions (A, B and C) are shown in Fig. 4. To establish it, the height of the image was divided into 19 strips 1 mm wide and the number of particles contained in each band

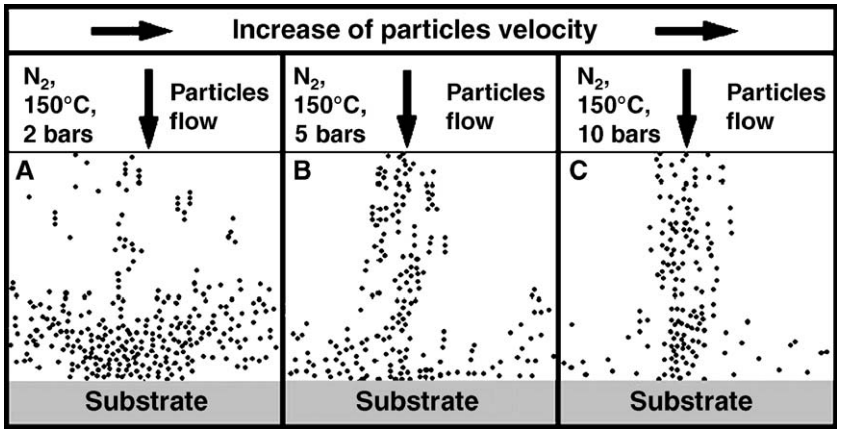

Fig. 3. Images of particle spray jet impacting on the substrate under CGDS conditions. Copper particles sprayed on copper substrate.

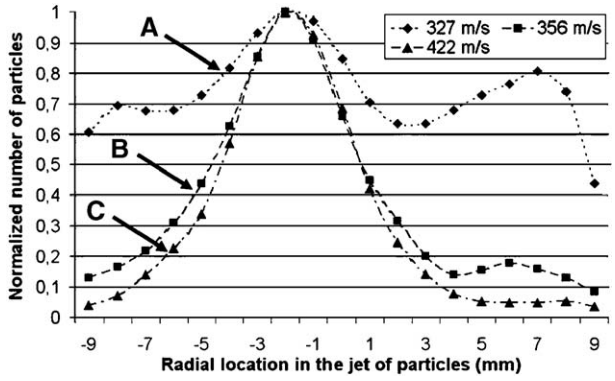

Fig. 4. Radial distribution of the particles impacting and rebounding on the substrate.

was counted and normalized with regard to the highest number of particles. Fig. 4 shows two types of curves:

- a rather large curve that corresponds to the spray conditions A for which the particle spray jet is large and most of the particle have an impact velocity below the critical one.
- Gaussian curves that correspond to the spray conditions B and C.

In the zone that stretches from 5 to 9 mm from the jet axis, the "humps" of curves show the particle rebounds. The height of these humps is representative of the number of particles that rebound on the surface. To try to quantify them, the following procedure is followed: subtraction of the value corresponding to the height of the hump and fitting of the new curve by a perfect Gaussian shape. The latter operation results in increase in the maximum value of the curve. The curve amplitude is found to be inversely proportional to the number of particle rebounds.

Fig. 5 shows the profiles of the curves after the application of this procedure and the corresponding deposition efficiency (DE). A difference between the curves corresponding to DE higher than zero and the curve corresponding to DE equal zero is now noticeable. When the maximum of the normalized amplitude is higher than 0.9, the particles start to stick on the substrate and therefore have reached a velocity higher than the critical one.

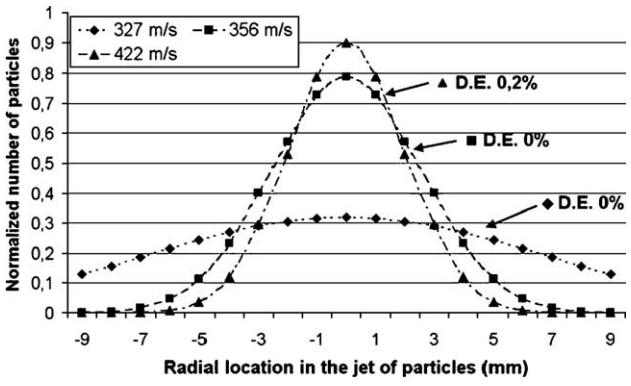

Fig. 5. Radial distribution of particles after curve fitting and corresponding deposition efficiency.

Table 3
Critical velocities determined by using the imaging technique

Powder	Substrate	Literature [2]	Measurement	Measurement standard deviation
Cu	Cu	571 m/s	422 m/s	45 m/s
Cu	316L	574 m/s	437 m/s	47 m/s
Ni	Cu	576 m/s	512 m/s	59 m/s

However, the velocity found in the conditions of the study (422 m/s) is lower than that found in the literature (571 m/s). Therefore, it is necessary to carry out further experiments with various materials sprayed on various substrates to validate the relevance of the method.

3.3. Copper and nickel powders sprayed on various substrates

Experiments have been carried out with copper powder sprayed on stainless steel (316L) and copper substrates and nickel powder sprayed on copper substrates. The results are summarized in Table 3. They showed that the transition from zero to positive deposition efficiency corresponds to amplitude of 0.9. They also showed that the decrease in the width of the Gaussian curve is not necessarily an indication in an increase in D. E.

The critical velocities determined by this method are lower that the ones found in the literature [2]: the difference ranges between 12 and 25%. Therefore a question arises from this comparison: does the measured velocity correspond to the velocity of particles that have stuck to the substrate?

The size of the particles that form the coating when the spraying conditions correspond to those that lead to critical velocity was, then, estimated by using an image analysis technique. The cross section of the coating was polished with a diamond past (up to 1 μm grain size) and etched to reveal its microstructure. The area in this cross section, corresponding to each grain was estimated and converted into an equivalent diameter which corresponds to the diameter of a circle having the same area than the grain. Several cross sections analysis in various orientations showed that the deformation was roughly similar, and allowed an estimation of the grain size in the first layer. The results revealed that the coating was mainly formed by the finest particles of the powder batch. For instance, for the copper coatings, the mean size of the particles in the coating was about 6.5 μm while the mean size of the original particles was 16.2 μm.

Therefore, it seems that the particles that stick on the substrate are too small to be correctly detected by the measurement technique as the light signal emitted by these particles can be smaller than the detection threshold.

Table 4
Critical velocities estimated by using the imaging technique and corresponding predictions

Powder	Substrate	Literature [2]	Measured critical velocity	Standard deviation	Calculated
Cu	Cu	571 m/s	422 m/s	45 m/s	520 m/s
Cu	316	574 m/s	437 m/s	47 m/s	523 m/s
Ni	Cu	576 m/s	512 m/s	59 m/s	582 m/s

The one-dimensional isentropic theory (1) makes it possible to roughly calculate the velocity of the particles under given spraying conditions [6,7].

$$v_p = M_a\sqrt{\gamma RT}\sqrt{\frac{C_D A_p \rho_g x}{m_p}} \quad (1)$$

Where M_a is the Mach number of the gas, γ is the ratio of heat capacity of gas at constant pressure to that at constant volume; R is the specific gas constant, C_D is the drag coefficient, A_p is the area of the particle, ρ_g is the density of the gas, x is the distance covered by the particle inside the nozzle and m_p is the mass of the particle.

The results of the experiments and calculations are summarized in Table 4.

The predicted velocities are closer to the ones found in the literature. However, it can also be noticed that the critical velocity of nickel determined by the imaging technique is closer to the predicted ones than that determined for copper. This could be explained by a smaller quantity of fine powder in the nickel powder. Indeed, the particles that started to stick to the substrate were about 7 μm in diameter.

Since this study deals with an actual material exhibiting a specific particle size range and mechanical properties, it is not surprising to find a difference of about 9% between the measured and predicted values.

4. Estimation of the critical velocity from the deposition efficiency

The deposition efficiency gives an indication about the amount of particles that have an impact velocity at least equal to the critical velocity. Assuming that the shape of the particles size curve (Fig. 6) is equivalent to the shape of the D.E. curve, the D.E. value can be directly linked to the diameter of the largest particle which can stick onto the substrate. This specific particle has a velocity equal to the critical one (Fig. 6). Since the spraying parameters (nature of the propellant gas, gas stagnation pressure and temperature) are known, the critical velocity of this particle can be calculated using the one-dimensional isentropic Eq. (1).

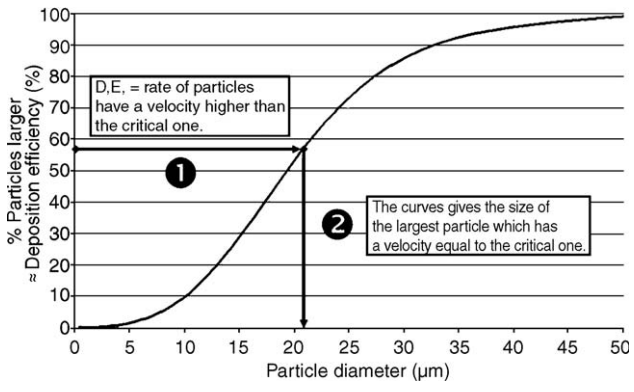

Fig. 6. Determination of the diameter of the particle that has a velocity equal to the critical one.

Table 5
Critical velocities calculated by using the one-dimensional isentropic equations

Powder	Substrate	Literature [2]	Imaging technique	Predictions from D.E.
Cu	Cu	571 m/s	520 m/s	538 m/s
Cu	316	574 m/s	523 m/s	555 m/s
Ni	Cu	576 m/s	582 m/s	585 m/s

To obtain the values of the critical velocity with a good accuracy, it is better to carry out several experiments and use the average value.

The results are summarized in Table 5.

The values found with this technique are in good agreement with the values found in the literature and with the imaging technique (after correction of the detected particle size).

5. Discussion

The various cases studied in this work validate the assumption that the critical velocity can be determined by quantifying the number of rebounds on the substrate. The key parameter of this method appears to be the amplitude of the curve corresponding to the radial distribution of particles in the spray jet. If this amplitude reaches the value of 0.9, it is an indication that the smallest particles start to stick on the substrate. The Gaussian shape of the curve also indicates that the particles which bounce off in the periphery of the jet represent less than 10% of the particles impacting in the jet center (Fig. 5).

The observation of the onset of coating formation is easier to carry out than the measurement of the velocity of the particles that effectively stick on the substrate. The 1-D mathematical model used to calculate the particle velocity shows that the predicted critical velocity corresponds to the velocity of the particle which has the mean diameter. Particles able to stick are about two times smaller and are faster. Velocities calculated for the operating condition leading to the critical velocity (temperature, pressure and size of particle which bound) are closer to the expected one. This is particularly true for the nickel powder probably due to its smaller grain size range.

The critical velocity calculated in the case of copper powder is about 9% lower than the one found in the literature. This difference can be attributed to various reasons: (i) The actual powder exhibits particle size distribution contrary to the powder of the model in which particles have the same size. (ii) The particles are not perfect spheres. (iii) The mechanical properties (hardness, Young modulus…) of particles can be different from that of the bulk material.

This work also shows that the critical velocity can be calculated by using the deposition efficiency of the spraying process. This second method gives results consistent with the values found in literature.

6. Conclusion

The change of the impact particle velocity for critical velocity can be detected by counting the number of rebounds on the substrate. The system described in this paper makes it possible to

simultaneously measure particle velocity and number of particle rebounds. However, the lack of brightness of the particles which have a velocity exceeding the critical one leads to disturb the measurement of velocity. Detected particles are mainly the coarser ones and so measured velocity is far lower than the velocity of particles which have really reached the critical one. The particle velocities calculated with the one-dimensional isentropic model under the same operating conditions are close to the values found in the literature. The difference between predicted and experimental values arises from the use of real powders in the experiments and not ideal ones as in the model.

Using the one-dimensional isentropic theory and value of deposition efficiency also makes it possible to estimate the critical velocity with a good accuracy. The results are consistent with those found with the imaging technique.

Finally, to improve the method based on the imaging technique, the accuracy of measurements carried out on small particles must be improved. Also, using batches of powder with a low content of fine particles will help to refine the experimentally-determined critical velocity for a given set of spraying conditions.

References

[1] Alkhimov, Kosarev, Papyrin, Al, Gas dynamic method for applying coating; US patent 5 302 414; 04/12/1994.
[2] Grujicic, Zhao, DeRosset, Helfritch, Mater. Des. 25 (2004) 681.
[3] Assadi, Gartner, Stoltenhoff, Kreye, Acta Mater. 51 (3) (2003) 4379.
[4] Largo, Hemline, Erika, in: C. Moreau, B. Marple (Eds.), Advancing the science and applying the technology, ASM Int., Materials Park, Ohio, USA, 2003, p. 1113.
[5] Vattulainen, Hämäläinen, Hernberg, Al, J. Therm. Spray Technol. 10 (1) (2001) 94.
[6] Dykhuizen, Smith, J. Therm. Spray Technol. 7 (2) (1998) 205.
[7] Jodoin, J. Therm. Spray Technol. 11 (4) (2002) 496.