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Effect of Substrate Temperature on the Formation Mechanism of Cold-Sprayed Aluminum, Zinc and Tin Coatings

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When describing the cold-spray process, one of the most widely used concepts is the critical velocity. Current models predicting critical velocities take the temperature of the sprayed particles explicitly into account, but not the surface temperature (substrate or already deposited layers) on which the particle impacts. This surface temperature is expected to play an important role, since the deformation process leading to particle bonding and coating formation takes place both on the particle and the substrate side. The aim of this work is to investigate the effect of the substrate temperature on the coating formation process. Experiments were performed using aluminum, zinc, and tin powders as coating materials. These materials have a rather large difference in critical velocities that gives the possibility to cover a broad range of deposition velocity to critical velocity ratio using commercial low-pressure cold-spray system. The sample surface was heated and the temperature was varied from room temperature to a high fraction of the melting point of the coating material for all three materials. The change in temperature of the substrate during the deposition process was measured by means of a high speed IR camera. The coating formation was investigated as a function of (1) the measured surface temperature of the substrate during deposition, (2) the gun transverse speed, and (3) the particle velocity. Both single particle impact samples and thick coatings were produced and characterized. Both the particle-substrate and interparticle bonding were evaluated by scanning electron microscopy (SEM) and confocal microscopy.

Keywords adhesion mechanism, coating-formation mechanism, cold-gas dynamic spraying

1. Introduction

Cold-spray technology is the most recent of the thermal-spray family. It derives its name from the fact that in these techniques, the particles stay in the solid state, with the exception of some particle jetting occurring when high adiabatic shear instability are developed around the particle surface during high velocity impact (Ref 1, 2). However, despite its name, the cold-spray technology requires some heating, mostly to reduce the propelling gas density, which increases the sound velocity in the heated gas and which enables to achieve higher velocities.

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The particles are also heated in contact with the hot gas affecting, therefore, their mechanical properties and, in some case, favoring the development of shear instabilities and the coating formation. Commercial cold spray systems heat the gas to temperature up to 500-800 °C depending on the specific system. Theoretical models (Ref 3), taking into account the particle temperature in the determination of a critical particle velocity, predicted that the critical velocity will decrease by 40 m/s for a 100 °C increase in the particle temperature. Lee et al. (Ref 4), showed experimentally that the critical velocity decreases by 50 m/s as the process gas temperature increases by 100 °C. All those results confirm the importance of the temperature in cold spray. However, focusing only on the particle temperature, those studies neglected another important side of the coating formation mechanism: the substrate temperature.

The scope of this article is to investigate the effect of the substrate temperature on the coating formation process.

2. Experimental

2.1 Coating Production

Coatings were produced using a low-pressure cold-spray system (SST, Centerline, Ontario, Canada). This system can operate either with air or with nitrogen at

Table 1 Spray conditions, T_{gun}, T_{pre}, and T_{imp} are the temperatures of the gun, the temperature of the preheated substrate and the measured temperature at the impact point of the spray jet respectively

T_{gun} (°C)	T_{pre} (°C)	T_{imp} (°C)	P (psi)	Vt (mm/s)	V (m/s)
Al					
33	33	…	90	2	397
200	33	…	90	2	475
400	33	…	90	2	568
500	33	219	90	2	615
500	130	270	90	2	615
500	230	290	90	2	615
500	330	302	90	2	615
Zn					
33	33	…	90	2	373
200	33	…	90	2	423
400	33	…	90	2	462
500	33	219	90	2	502
500	105	263	90	2	502
500	170	280	90	2	502
500	245	292	90	2	502
Sn					
33	33	33	90	2	…
200	33	74	90	2	…
33	80	45	90	2	…
200	80	100	90	2	…

pressure typically between 0.31 and 0.62 MPa For the present study nitrogen was used at 0.62 MPa, the nitrogen can be heated in the gun region up to 500 °C. The powders are injected radially into the high-velocity air jet taking advantage of a low-pressure area near the throat. In order to insure a constant particle feed rate, a thermal-spray powder feeder was used instead of the included powder feeder. The powder gas flow rate of 3 SLPM was used to ensure proper feeding. A gun transverse velocity of 2 mm/s was used at a stand off distance of 10 mm unless otherwise specified. The spray conditions are listed in Table 1.

The coatings were produced on flat 3 mm thick grit-blasted carbon-steel coupons. In order to investigate the effect of surface temperature, a heated specimen holder was used. The holder was made of (15 mm thick) stainless steel plate into which three heating cartridges were inserted (5 mm diameter 1200 W). This configuration enables a uniform heating of the specimen from room temperature up to 350 °C. Up to this temperature weight gain was measured both on bare and coated substrate after being heated in air for 1 h.

2.2 Particle Flow Characterization

The particle flow was characterized using a DPV-2000 system (Tecnar Automation, St-Bruno, QC, Canada), adapted for the monitoring of cold particles. A laser diode (7 W, $\lambda = 830$ nm) was used to illuminate the in-flight particles. In order to maximize the signal to noise ratio, a 830 nm filter with a bandwidth of 10 nm was used. The use of laser illumination enables the detection of particles in absence of sufficient thermal emission. The sensor head was positioned to collect the light scattered at 10 degrees from the laser direction. The distance between the center of the particle jet and the sensor head was 10 cm.

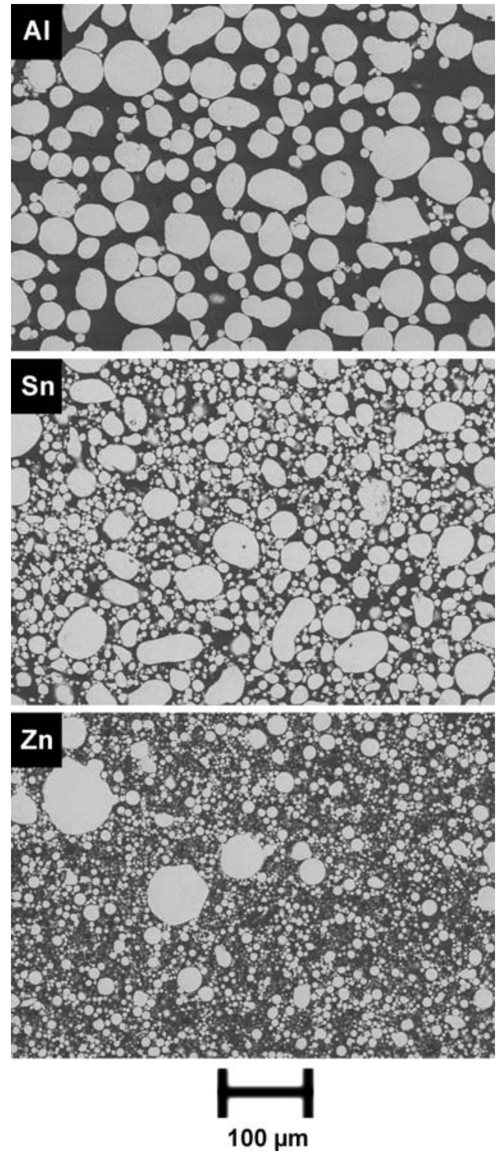

Fig. 1 Cross-section micrographs of the Al, Sn, and Zn powders

The forward scattering geometry was used to minimize the influence of the particle shape on the measured particle diameter and to maximize the scattering light signal. Since the monitored particles were relatively spherical, the measured size distribution is expected to be accurately represented in the presented case.

2.3 Materials

Three materials were used: Al, Zn, and Sn (Alpha Aesar). The cross-section micrographs of powders and the particle size distributions are presented in Fig. 1 and 2, respectively. The average particle size was found to be 36.2, 13.6, and 10.6 μm for Al, Sn, and Zn, respectively. It can be noticed that the size of Al powder is similar to that of the largest particles of both Zn and Sn powders, while the Zn powder contains the largest number of small particles.

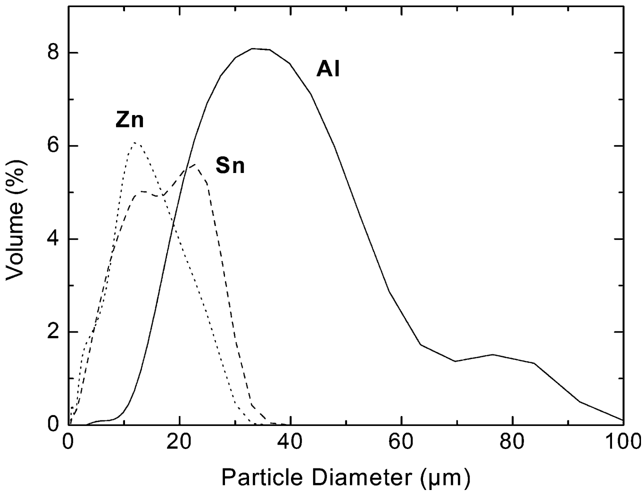

Fig. 2 Particle size distributions of the Al, Sn, and Zn powders

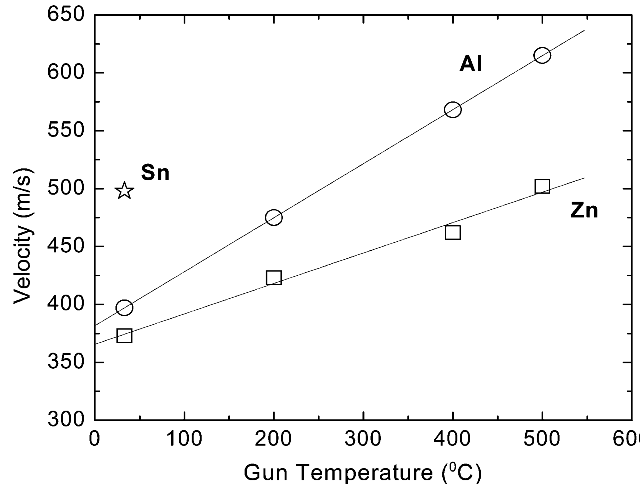

Fig. 3 Particles velocity vs. gun temperature relationship for Al and Zn powders. Constant spray distance of 1 cm and nitrogen pressure of 0.62 MPa

2.4 Surface Temperature

The surface temperature of coatings during deposition was measured by means of an infrared high-speed camera (Thermacam SC3000) recording thermal emission from the sample surface during deposition. The thermal images recorded at a rate of 60 images per second enable the monitoring of the temperature on specific locations, such as the impact point of particle jet for example. The camera software enables detail data analysis, making it possible to take into account specific areas and assigned emissivity values.

2.5 Coating Microstructure

Coatings microstructures were characterized using SEM after standard vacuum infiltration and polishing procedure on the samples.

3. Results and Discussion

3.1 Gun Characterization

The Al and Zn average particle velocity was measured for different gun temperatures ranging from 33 to 500 °C. The results are presented in Fig. 3. The relationships are linear, as predicted by fluid dynamic calculation (Ref 5, 6). Due to its low melting temperature, similar measurements were not performed on Sn. The velocity of Sn was however measured at a gun temperature of 33 °C and the results are shown in Fig. 3.

3.2 Substrate Characterization

The effect of the gun temperature on the substrate temperature was investigated using infrared imaging. Series of films were taken for the different spray conditions on heated and nonheated substrates. Figure 4 presents example of images taken from some of these films. It can clearly be seen that nonpreheated samples can be heated significantly by the supersonic impacting gas jet (Fig. 4a to d). Controlling the substrate preheating temperature makes it possible also to change the temperature of the substrate surface without, however, modifying the particle impact velocity. Figure 4e to h shows the influence of the substrate preheating on the surface temperature distribution during spraying.

Quantitative analysis of the recorded thermal images enables the dynamic follow-up of the surface temperature while the gun transverses the parts. Figure 5 presents the maximum surface temperature (T_{imp}) of the substrate in an area crossed by the gas jet while moving at 2 mm/s. As seen in Fig. 5, a linear relation was found between the gun temperature (T_{gun}) and the impact temperature (T_{imp}).

3.3 Effect on Deposition Efficiency

Deposition efficiency has been measured as the ratio of the sample weight increase over the mass of powder injected into the gun during the time that the gun is actually over the sample. Figure 6(a) presents the deposition efficiency, as a function of the gun temperature. From this graph for both the aluminum and the zinc powders the efficiency increases with the increase in gun temperature. It is also possible to express the deposition efficiency as a function of the particle velocity (Fig. 6b). It can be seen that the deposition starts to be measurable for average velocities of about 400 and 500 m/s for Zn and Al, respectively. These values are inferior to those reported in the literature (Ref 7-9). This difference can be understood by the fact that particle velocities are distributed over a range of value and that only a fraction of them participate to the coating formation as testified by the low measured deposition efficiency. For the Sn powder no significant deposition efficiency was obtained for the tested conditions.

As shown in Fig. 4 changing the gun temperature modifies not only the particle velocities, but also the

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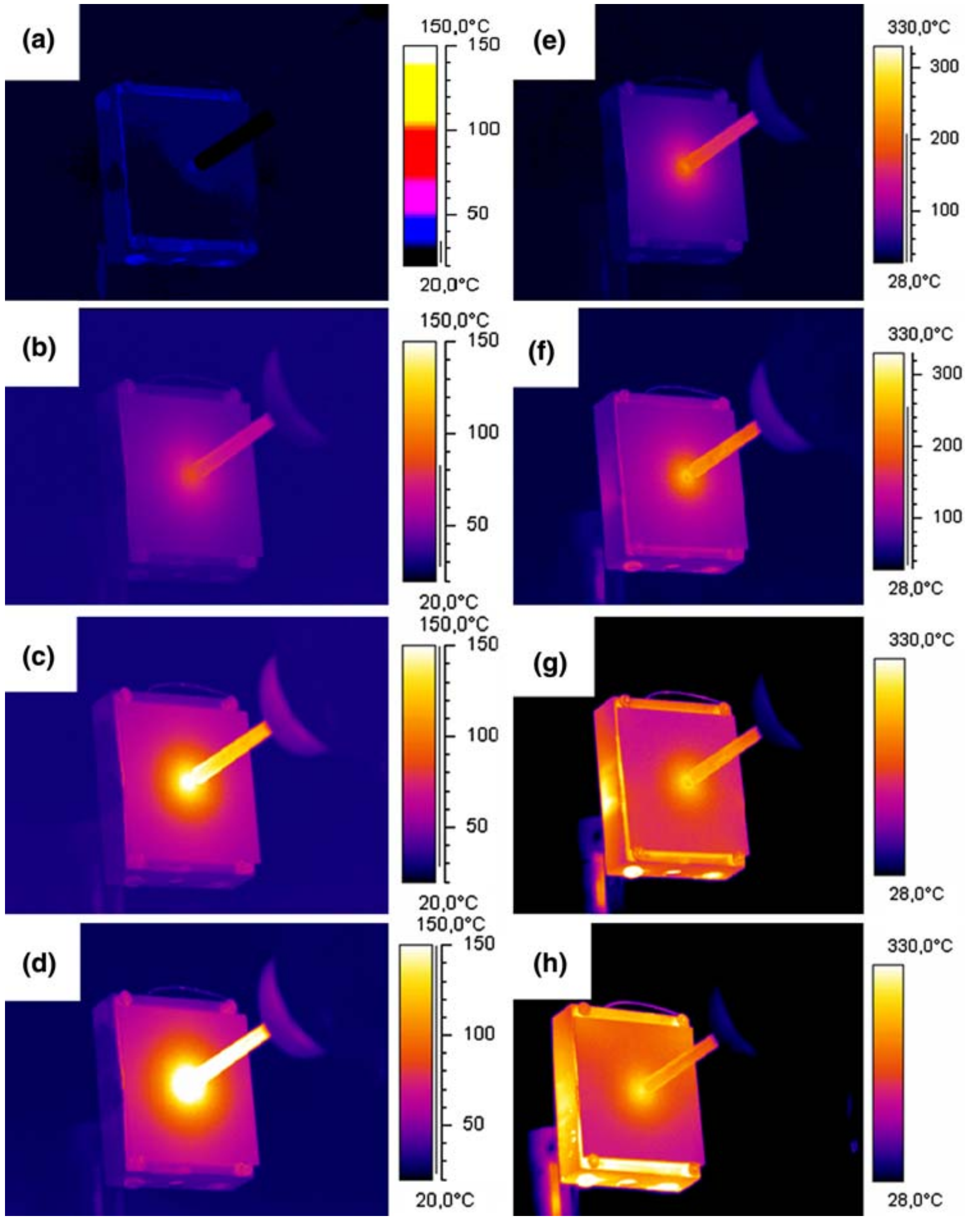

Fig. 4 Infrared images taken during spraying in various conditions: *T* substrate 33 °C, *T* gun inlet 33, 200, 400, and 500 °C, (a, b, c, and d), respectively. *T* gun inlet 500 °C, temperature of substrate 33, 105, 245, and 330 °C, (e to h)

surface temperature. In order to isolate the effect of T_{imp} on the deposition efficiency, experiments using the same spray conditions (gun temperature of 500 °C) for different substrate preheating temperature were undertaken. Figure 7 presents the variation of T_{imp} as a function of T_{pre} for the gun temperature of 500 °C. The impact temperature can be adjusted between 220 and 305 °C as the substrate is heated from 33 to 330 °C. It should be noted that for the highest preheating temperature the impact point is cooled down by the impinging gas jet.

Results of the deposition efficiency measurement as a function of the surface temperature are presented in Fig. 8. It can be seen that the zinc deposition efficiency decreases with temperature. On the other hand it increases for Al and it remains almost zero for tin.

When comparing material properties as a function of temperature it is often useful to express this temperature as the ratio of the process temperature (T_{imp}) to the material melting temperature (T_m) in Kelvin. Figure 9 presents the deposition efficiency as a function of the ratio T_{imp}/T_m. Since of the difference in melting point of the different materials the Zn and Al curves are not superimposed anymore. Interestingly the Zn curve decreases from T_{imp}/T_m of 0.7 while the Al curve increases up to a ratio of temperature of 0.6. These results indicate potential optimum surface temperature ratio between 0.6 and

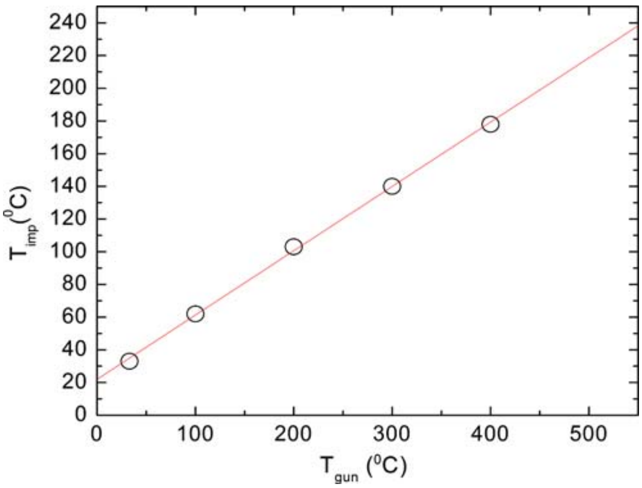

Fig. 5 Effect of gun temperature on the surface temperature

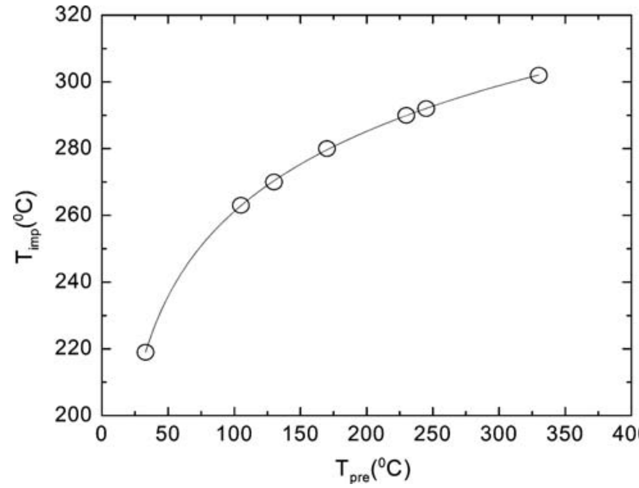

Fig. 7 Temperature at the jet impact point as a function of the preheated temperature for a gun temperature of 500 °C

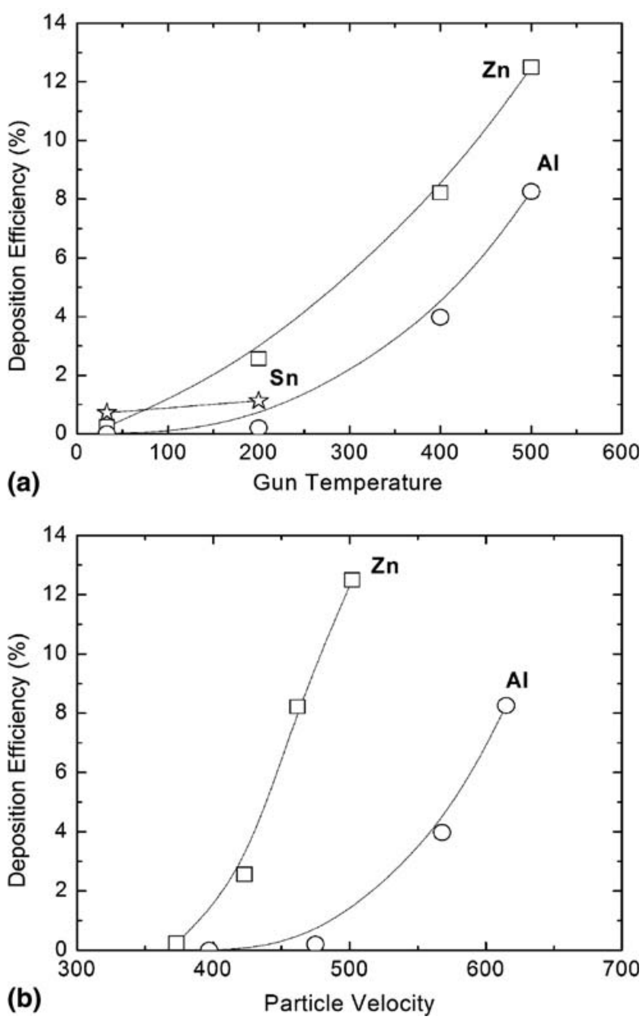

Fig. 6 Deposition efficiency as a function of gun inlet temperature (a) and the average particle velocity (b)

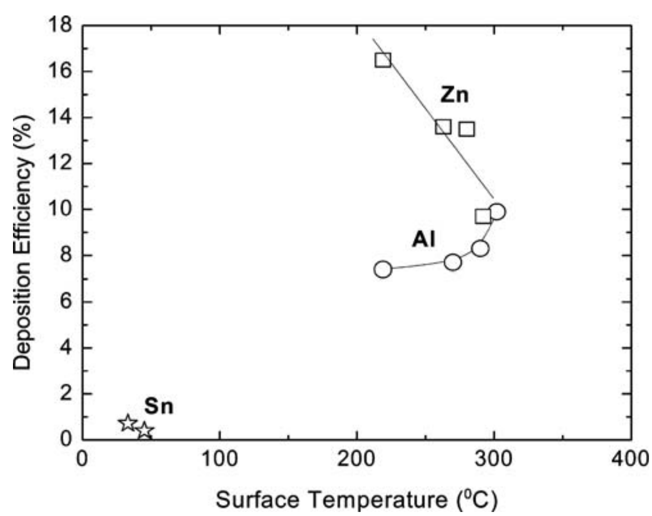

Fig. 8 Deposition efficiency as function of substrate temperature

0.7. However, in this range of temperature ratio is at which the Sn powder showed always very low deposition efficiency. This is indicating that for this material another phenomena are influencing the coating formation, a more detailed discussion of the tin results will be presented later.

3.4 Microstructure

Figure 10 presents the microstructures of Zn and Al coatings produced under different spray conditions. For the aluminum coating at low particle velocity (397 m/s) on cold substrate (33 °C) (Fig. 10a) only some Al particles entrapped in the sandblasted substrate surface asperities are visible. It should be noted that the top surface of these embedded particles is relatively flat indicating that they have been hammered by other particles. These particles

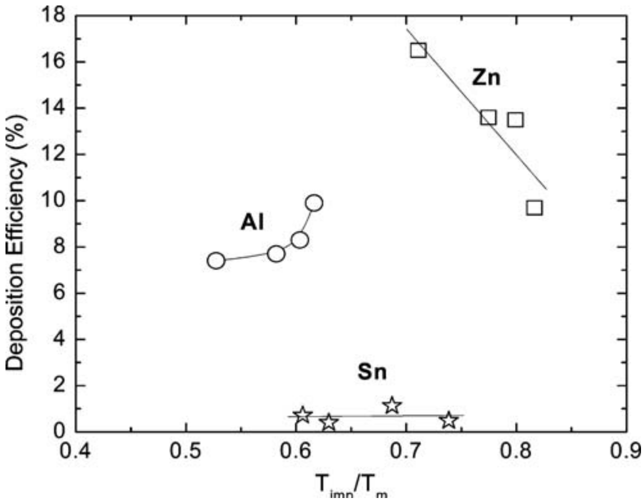

Fig. 9 Deposition efficiency as a function of the ratio of the surface temperature to the melting temperature

were however unable to make a bond to the previously deposited material. With the increase in velocity and temperature (Fig. 10b and c) the coating thickness increased significantly and the interfacial region remained very clean, as it is difficult to see particle boundaries. For the Zn coatings, at low temperature (Fig. 10d) the few particles seen on the surface looks elongated and heavily deformed however they did not adhere well on the steel surface. For coatings produced at higher velocity the particle boundaries are clearly visible probably because of some particle oxidation occurring on the Zn particle's surfaces, the impact temperature being close to the melting temperature. For the coating produced without preheating (Fig. 10e) the particles seems less deformed with less particle contact, as compared to the coating produced with the substrate preheated at 330 °C (Fig. 10f).

3.5 Tin Coatings

Trying to understand the constant but low deposition efficiency for tin coating, the deposition efficiency was

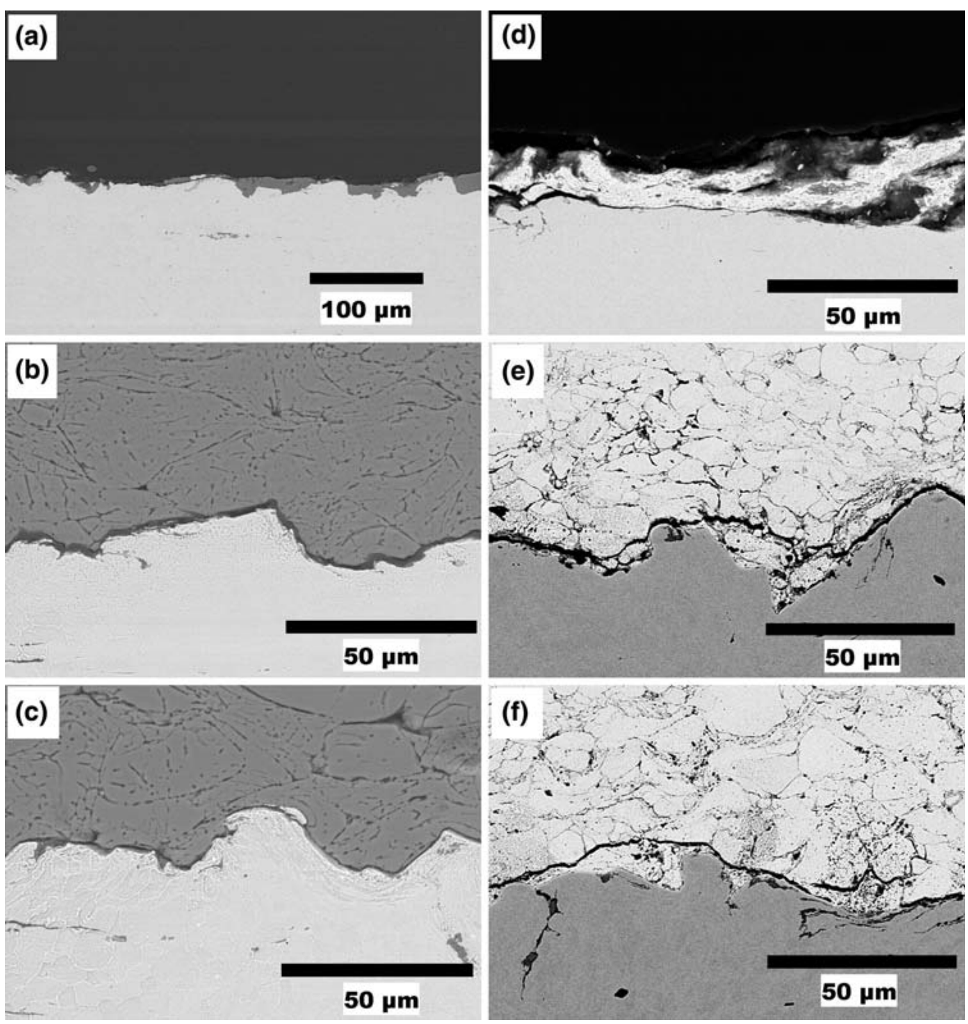

Fig. 10 Microstructure of Al (a, b, and c) and Zn (d, e, and f) coatings deposited on non preheated steel substrate at gun temperature of 33 °C (a and d); at 500 °C (b and e); at gun temperature of 500 °C for preheated temperature of 330 and 245 °C (c and f respectively)

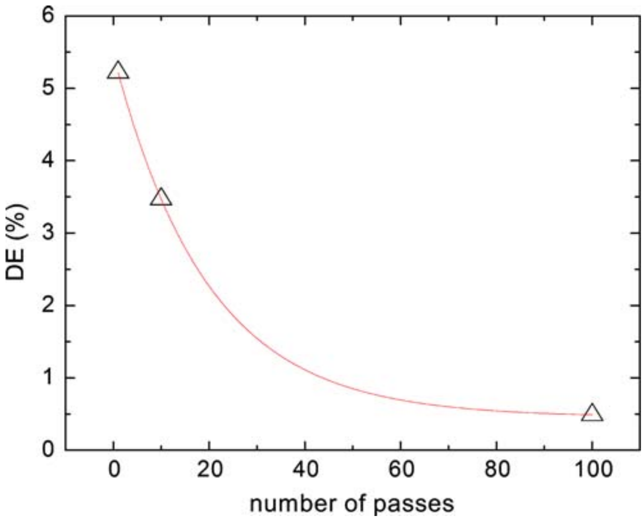

Fig. 11 Deposition efficiency of Sn as a function of the number of passes (T_{gun} = 33 °C, P = 0.6 MPa)

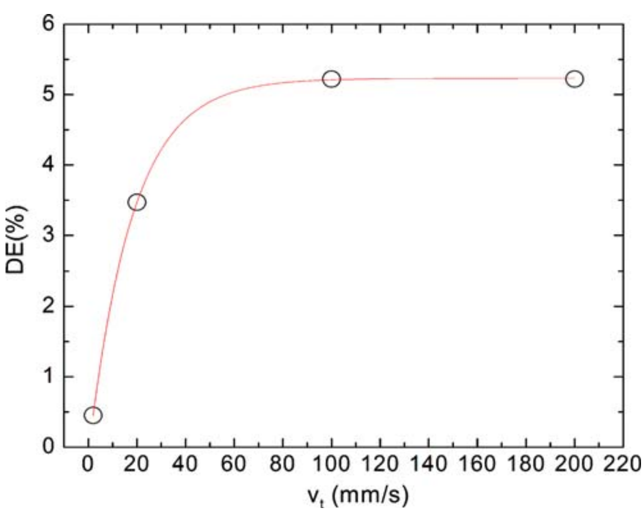

Fig. 12 Deposition efficiency as a function of the gun transverse speed (T_{gun} = 33 °C, P = 0.6 MPa)

measured as a function of the number of passes. It can be seen in Fig. 11 that the deposition efficiency decreases with the number of passes for the Sn coating. This indicates that a coating was formed only on the steel substrate and no further deposition was obtained on a Sn surface.

In order to vary the proportion of particles impacting a fresh steel surface, measurements were made varying the gun transverse speed and the results are presented in Fig 12. The deposition efficiency increases with the gun transverse speed and reach a maximum for velocity higher than 100 mm/s. This result can be interpreted by the fact that increasing transverse velocity increases the fraction of particles that hit the steel substrate. If the velocity is increased over a certain threshold than most of the incoming particles hits the steel substrate and no further improvement in DE can be obtained.

Micrographs of Sn samples produced at different gun transverse speed are presented in Fig. 13. It can be noticed that for 2 and 10 mm/s the steel surface was completely covered, while some uncoated areas are observed for 100 mm/s.

At 200 mm/s a large fraction of the surface remained uncoated. This correlates well with results in Fig. 12 where the maximum value of DE is obtained for velocities close to 100 mm/s; above this velocity there is always some fresh steel surface remaining exposed.

In order to understand better this behavior, isolated particles were impacted on polished steel and tin substrate. When sprayed on polished steel substrate no substrate deformation or particles adhesion was observed. On the contrary, when impacting on a sandblasted steel surface, many Sn particles can be found impaled on the surface asperities.

On polished Sn surface (Fig. 14) the Sn particles created craters during their impact but very few get stuck and adhere to the surface. This is in agreement with the fact that, in these experiments, the velocity of the particles is higher that the erosion velocity. The measured average velocity of the Sn particles was around 500 m/s, while the erosion velocity (i.e., the maximum velocity before the adiabatic shear instability becomes too important for the particle to bond to the surface) was calculated to be around 180 m/s for 25 micron particles by Schmidt et al. (Ref 7).

The behavior of Sn compared that of Zn and Al illustrates the existence of several important velocities describing the material behavior. At least two critical velocities can be defined: the first one is related to the bonding of particle on a substrate surface (this velocity was reached for Al and Sn deposited on steel while for Zn deposition on steel occur only for velocities higher that 375 m/s), the second critical velocity is related to the self bonding capability of a material and thus to the possibility to build thick coating (this velocity was reached for most of Al and Zn). Finally a third important velocity was also reached which is the erosion velocity that was reached for the Sn. A more detailed description of these velocities and their relationship with material properties and particle size can be found in Ref (9).

4. Summary

This study shows that surface temperature at the impact point of a low-pressure cold-spray system can be significantly influenced by the process gas temperature. The deposition efficiency changes as a function of the process gas temperature because of the influence of this processing parameter both on the velocity and on the surface temperature. At constant gun temperature, the surface temperature was varied using a substrate heater. For Al the deposition efficiency increases as the surface temperature was increased. For Zn it was the opposite, the DE decreases and for Sn no significant change was observed. For low melting point material such as Zn the

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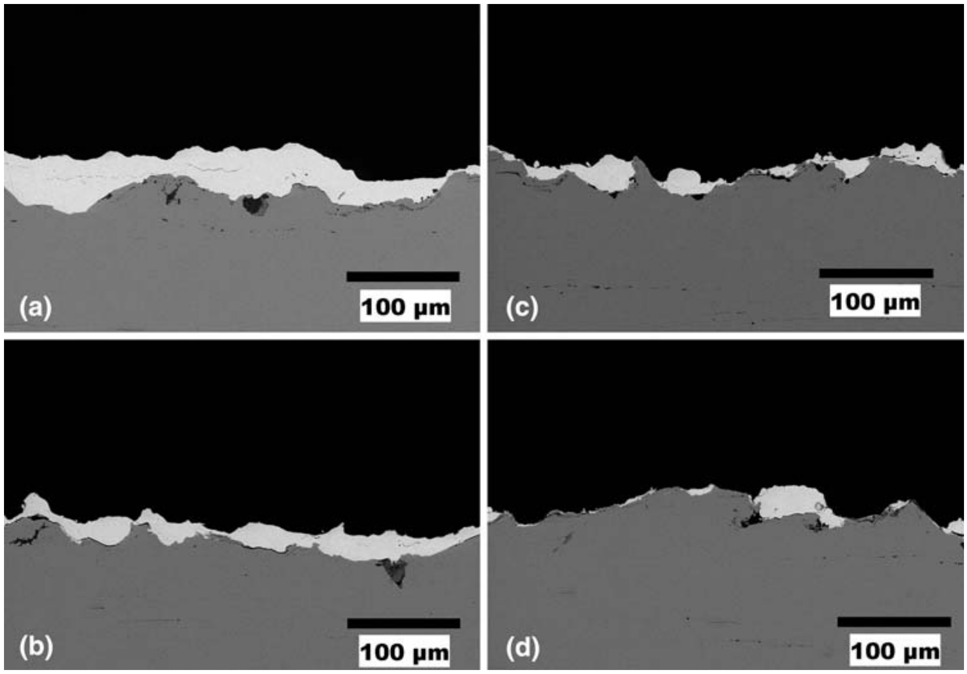

Fig. 13 Surface of tin coatings on steel surface with gun transverse velocity of: 2, 10, 100, and 200 m/s for (a), (b), (c), and (d) respectively (T_{gun} = 33 °C, $P = 0.6$ MPa)

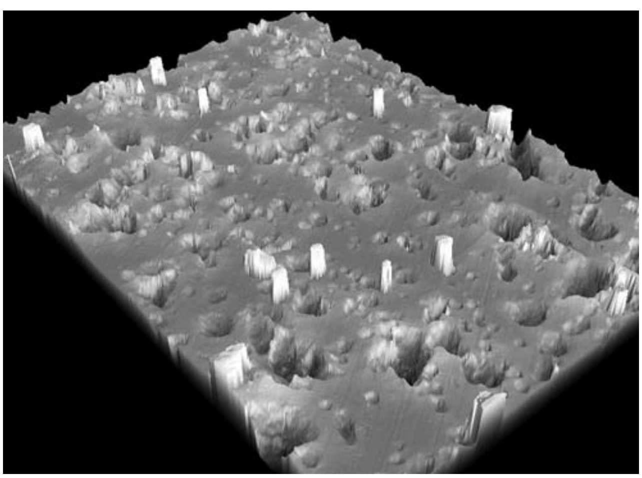

Fig. 14 3D view of particle impact on a pre polished tin samples substrate and gun temperature 33 °C gun pressure 90 psi

surface temperature might be too high compromising the deposition efficiency. For soft and low melting point material such as Sn, lower deposition efficiency was measured mainly because of the low self bonding property of the tin leading to a single particle layer formation of tin on the steel surface. The velocity of Sn was significantly higher than the predicted erosion velocity of Sn on Sn substrate and evidence of such erosion was demonstrated.

References

1. R.C. Dykhuizen, et al., Impact of high velocity cold spray particles, *J. Therm. Spray Technol.*, 1999, **8**(4), p 559-564
2. F. Gartner, et al., Numerical and Microstructural Investigations of the Bonding Mechanisms in Cold Spraying, *ITSC 2003: International Thermal Spray Conference*, ASM International, 2003
3. H. Assadi, et al., Bonding Mechanism in Cold Gas Spraying, *Acta Mater.*, 2003, **51**(15), p 4379-4394
4. J.H. Lee, et al., Effect of Particle Temperature on the Critical Velocity for Particle Deposition by Kinetic Spraying, *ITSC2006: International Thermal Spray Conference*, Seattle, 2006
5. B. Jodoin, F. Raletz, and M. Vardelle, Cold Spray Modeling and Validation Using an Optical Diagnostic Method, *Surf. Coat. Technol.*, 2006, **200**(14-15), p 4424-4432
6. B. Jodoin, Cold Spray Nozzle Mach Number Limitation, *J. Therm. Spray Technol.*, 2002, **11**(4), p 496-507
7. T. Schmidt, et al., Development of a Generalized Parameter Window for Cold Spray Deposition, *Acta Mater.*, 2006, **54**(3), p 729-742
8. F. Gartner, et al., The Cold Spray Process and Its Potential for Industrial Applications, *J. Therm. Spray Technol.*, 2006, **15**(2), p 223-232
9. T. Schmidt, F. Gaertner, and H. Kreye, New Developments in Cold Spray Based on Higher Gas and Particle Temperatures, *J. Therm. Spray Technol.*, 2006, **15**(4), p 488-494