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The bond strength of Al–Si coating on mild steel by kinetic spraying deposition

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Abstract

Kinetic spraying (or cold gas dynamic spraying) works by accelerating small solid particles to supersonic velocities, and then impacting them onto a substrate. These high impact velocities, and low particle temperatures are the principal attributes of kinetic spraying technology. However, only recently has this technology's interfacial behavior, due to particle/substrate impaction, become well understood. In order to investigate the particle/substrate bond behavior, Al–Si feedstock was deposited onto mild steel, over a range of particle velocities; next, their respective coating bond strengths were measured by the stud pull coating adherence test. The effects of the particle velocity and the substrate surface roughness on the coating bond strength were presented, and a model of the particle/substrate bond generation was discussed in an effort to estimate the bond strength.
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Keywords: Kinetic spraying; Cold gas dynamic spraying; Bond strength; High velocity impact

1. Introduction

Kinetic spraying works by accelerating small solid particles to supersonic velocities, and then impacting them onto a substrate; where the critical particle velocity is key in characterizing the process. In these processes, dense coatings are produced without significant heating of the spray powder or substrate material, therefore the kinetic energy of the particles plays a major role in the behavior of impaction and deformation. After over a decade of development, kinetic spraying has been successful in depositing a wide range of pure metals, metal alloys, polymers, composites and nano-materials onto a variety of substrate materials [1–3].

The bonding mechanisms associated with high velocity impaction can be explained by shear instabilities caused from thermal softening, which in turn are caused by adiabatic heating during high strain rate deformation. Numerous experimental studies [4–6] have shown that particles require a minimum critical velocity in order to deposit onto a substrate, while also suggesting that higher impact velocities yield a better coating bond strength and lower coating porosity. From computer modeling, Assadi et al. [4] have provided an equation to estimate the critical particle velocity as a function of the feedstock material properties, such as density, melting point, ultimate strength, and initial particle temperature; but not interfacial bonding.

Until now, the interfacial reactions due to high velocity impaction have not been well understood; even though many different interfacial reactions have been documented by experimental and theoretical investigations. In early research interfacial melting was observed, but was proved not to be a dominant mechanism in the high-speed particle/substrate bonding [6,7]. Since most depositing in kinetic spraying occurred in the solid state, with a high interfacial pressure and large extents of plastic deformation; atomic length-scale phenomena, atomic diffusion, and surface adhesion were all considered to be the dominant bonding reactions. In Bolestal et al. [8] and Xiong et al.'s [9] studies, the boundary phase of the intermetallic compound was checked by XRD, and indicated that an interface boundary, which included atomic diffusion, occurred during the kinetic spraying process. Furthermore, Bolestal observed that the thickness of the film interface was 20–50 nm. However, in most kinetic spraying processes, particle impaction is completed within 0.1 μs, during

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which the atomic diffusion was not significant. For example, the thickness of an Al–Cu inter-diffusion (with an inter-diffusion coefficient of 10^{-15} to 10^{-14} m^2/s) layer in 0.1 μs is less than 1 nm; which suggests that the atomic diffusion may not be a dominant mechanism of kinetic spraying after all.

Clean surfaces and high contact pressures, which come from high-speed impaction and large plastic deformation along the interface, made the two contact surfaces mutually conforming so that surface adhesion could occur. Metallic bonding was observed in the coatings, and therefore surface adhesion was believed to play an important role in the particle bonding. In Grujicic et al.'s [6] computational analysis of the interfacial bonding in a cold-gas dynamic-spray process, nano/micro-scale material mixing and mechanical interlocking were both identified and used to explain the enhancement of interfacial bonding. These phenomena can be exemplified by the interfacial roll-up and vortices observed during high shear and high viscous flow along the interface. Grujicic's mechanism is useful in explaining the coating of two materials with weak repulsive or attractive atomic interactions, which is generally difficult to explain solely by surface adhesion.

The present study tried to clarify the effects of particle velocity and substrate surface roughness on the coating bond strength. In addition, a model, of the particle/substrate bond generation, used to estimate the bond strength during high velocity impact was discussed.

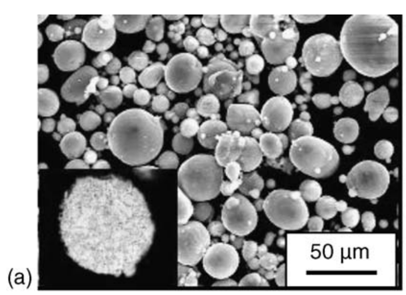
2. Experimental procedures

2.1. Spraying system

In this study, a commercially available CGT kinetic spraying system was used. The equipment and the coating process are described in detail in the literature [1–3]. A de Laval type nozzle with a converging/diverging inner form was used (standard nozzle type is when exit throat exit diameter ratio is 3.15). Nitrogen was used for both the process and feedstock carrier gas; where the pressure ranged between 0.3 and 3.0 MPa, and the temperature was fixed at 400 °C. The feedstock was Al–12Si powder with a mean particle size of 25 μm, a physical density of 2.66 g/cm^3, and a feed rate of 8 g/min. The micrograph and size distribution of the Al–Si powder feedstock are shown in Fig. 1. Mild steel was used as the substrate, and two kinds of surface conditions were prepared by either polishing or grit-blasting the surface. The roughness R_a of the as-polished surface and the grit-blasted surface were 1.52 and 17.98, respectively, which was measured with a laser scan microscope. Finally, the target substrate distance was fixed at 30 mm in front of the nozzle exit.

2.2. Analysis

During our experiments, the SprayWatch system (Oseir Ltd., Finland) was used to measure the velocity of in-flight particles. With this system, images of the flying particles were taken with a high-speed camera; then from the particle flying distance and camera exposure time a particle velocity could be calculated. While the particle size, distance to the nozzle exit, radial position, and other factors affected the particles' velocity; only the effect of the mean particle velocity was analyzed for the purpose of simplification. In an effort to capture the most accurate representation of the process, the mean particle velocity was determined from a large number (>300) of measured flying particles. The remaining experimental analysis was accomplished with optical microscopy, X-ray diffraction (XRD), and scanning electron microscopy (SEM).



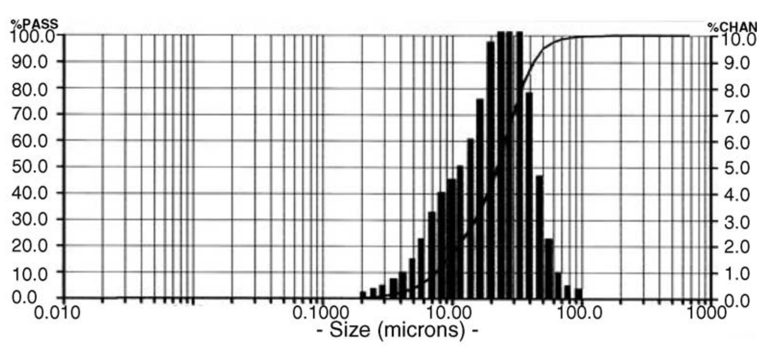


Fig. 1. Feedstock: Al–Si (wt.% 78:12), spherical, +5 to 45 μm (mean size 25 μm): (a) SEM micrograph of Al–Si powder morphology; (b) laser scatter value of Al–Si powder size distribution (volume fraction).

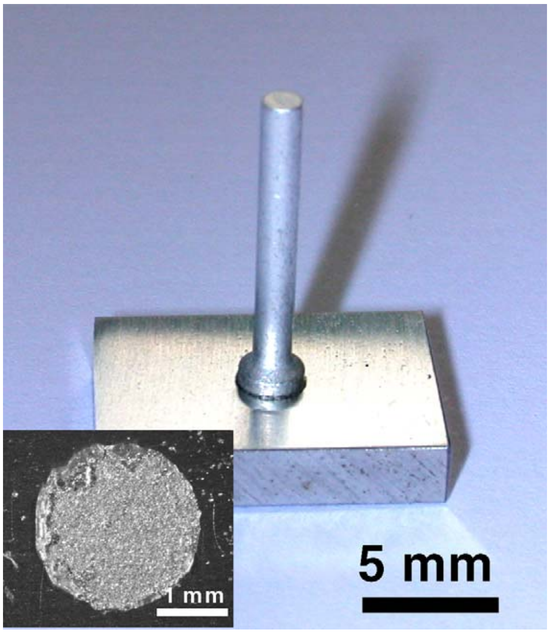


Fig. 2. Optical graphs of the Stud Pull Coating Adherence Test specimen and the fracture surface on the substrate after testing.

To measure the bond strength of coating/substrate, the Stud Pull Coating Adherence Test was carried out by using a Romulus Bond Strength Tester. The coating specimens were cut to 10 mm × 12 mm rectangles. Aluminum test studs with a 2.70 mm diameter head and 12.5 mm length, were attached to the coating surfaces, which was micro-polished with 0.3 μm alumina before, as shown in Fig. 2. The tests used a unique, ultra-strong, non-stressing, thermally curing, epoxy-bonding agent (with an ultimate strength higher than 85 MPa), which had been pre-applied to the face of the studs. Finally, the test stud assemblies were placed in an oven and cured at 150 °C for 90 min. After which a stud pull test could be conducted, and the bond strength evaluated. Romulus Bond Strength Tester is not a standard method for coating bond strength measurement, but it is a easy way to estimate the strengths of coating.

3. Results and discussion

3.1. Bond strength of coating/substrate

In our previous work [10], particle velocity and deposition efficiency of Al–Si powder under different gas conditions was discussed. The critical velocity for Al–Si feedstock deposition onto the mild steel substrate was found to be about 580 m/s, and full surface coatings were produced with impact velocities higher than this critical velocity. Specimens with coating layers thicker than 250 μm were used for the Stud Pull Coating Adherence Test. After the adherence test, the deposits were removed with the epoxy applied studs, thus leaving fragments on the fracture surface of the substrate, as show in Fig. 3; showing that the cohesive strength of the coatings is higher than the adhesive strength between the deposit and the substrate. The fragments left on the fracture surface can be thought to be the result of the cohesion reduction by the inner pores and defects in the coatings. The area frictions of fragments on the fracture surface for as-polished and grit-blasted specimens are about 10 and 2%, respectively. So, the bond strength measured in this work is a combination of adhesive and cohesive strength of Al–Si/mild steel assembly.

Fig. 4 shows the bond strength of the coatings under different particle velocities. It is observed that the measured bond strength of the kinetic sprayed coatings were moderately higher than those of thermal sprayed coatings [11–13], where most tensile bond strengths were lower than 30 MPa. For the Al–Si/mild steel assemblage in kinetic spraying, maximum bond strength of approximately 70 MPa is obtained. The full surface coating experiments were carried out with particle impact velocities ranging between 500 and 800 m/s. When the particle velocity is lower than the critical velocity (580 m/s), it is difficult to deposit the coatings or the coatings are too thin for bond strength test. In the case that the particle velocity is high enough for feedstock deposition, the bond strengths of 20–70 MPa are produced by increasing the particle impact velocity. This result allows us to deduce that the highest bonding strength is achieved through kinetic spray deposition; while the strongest adhesion is obtained under high particle impact velocities. The theory of particle deposition in thermal and

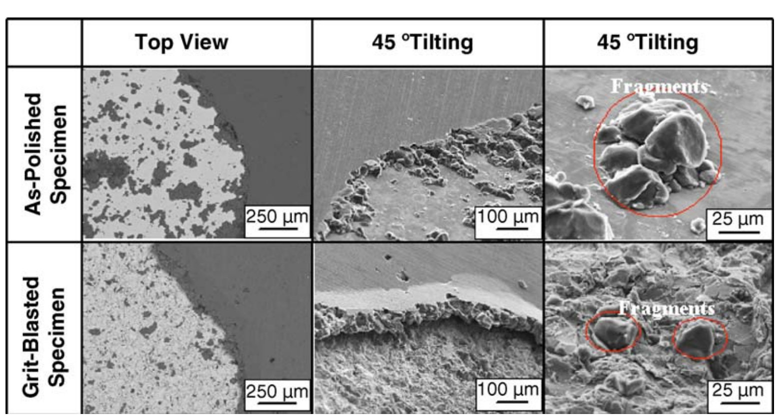


Fig. 3. SEM micrographs of the fracture surfaces on the substrates after the Stud Pull Coating Adherence Test.

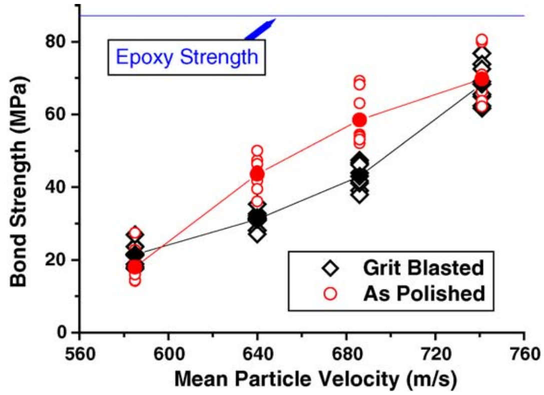


Fig. 4. Measured adhesive bond strength of Al–Si/mild steel coatings with different impact velocities.

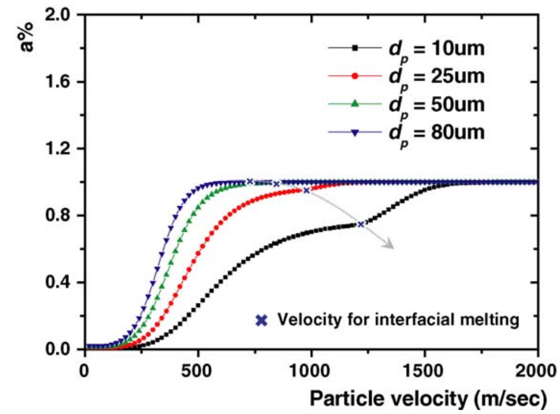


Fig. 5. Calculated fraction of bonded atoms for varisized Al–Si feedstock impacting onto the mild steel substrate. Here, "✖" indicates the minimum required velocities for interfacial melting.

kinetic spraying processes, assumes that the particle attaches onto the substrate when high velocity impaction is used to generate a bond between the two impact surfaces. Therefore, it is suggested that a higher fraction of bonded atoms, between two impacted surfaces, is produced when a higher impact velocity is applied.

3.2. Bond generation during high velocity impact

In the modeling of bond generation, during high velocity impaction, the key interaction is the adhesion of the feedstock particle to the substrate; which is characterized by the generation of bonds between the two contact surfaces. The adhesive strength can be expressed as: $\sigma = a\%\sigma_{\max}$, where $\sigma_{\max}$ is the maximum adhesive strength of a given particle to the substrate; $a\%$ is the fraction of bonded atoms per unit adhesive interface, and is also called the relative strength of the bond between particle and substrate. In the investigation into high speed thermal spraying interactions (D-Gun Spraying), Shorshorov and Kharlamov [14] developed a relation for calculating the fraction of bonded atoms. Kurochkin et al. [15] improved this relation for the kinetic spraying process and obtained the following expression:

$$a\% = 1 - \exp\left\{ -\nu t_c \exp\left[\frac{-E_a}{kT_c + (1-e_r)m_a v_p^2/2}\right]\right\}. \quad (1)$$

where ν is the natural frequency of eigen oscillations of atoms in the crystal lattice, t_c the contact time, E_a the activation energy of the chemical bonds ($E_a = 0.5 \times 10^{-19}$ J for pure aluminum and $E_a = 1.55 \times 10^{-19}$ J for iron [5]), T_c the contact temperature, k the Boltzman constant, e_r the recoil coefficient during elastic recovering, m_a the atom mass of the impact particle, and v_p the velocity of the impact particle. In Eq. (1), $a\%$ is mainly affected by the contact temperature T_c and impact velocity v_p.

Under high speed impaction, the particle/substrate interaction is considered an adiabatic process, where all heat transfer is ignored. From the thermal diffusion distance: $z = \sqrt{\chi_p t_c}$, where χ_p is the thermal diffusivity of particle material, and t_c the contact time, a heated region near the contact surface can be partitioned. Papyrin et al. [5] gave a simple estimation method for contact temperature, which is the average value when a uniform heating in the heated region is assumed. Alkhimov et al. [16,17] pointed out that the temperature near the contact boundary decreased with distance from the contact interface. If not for these estimations, the complete solution of contact temperature and temperature distribution would be very complicated for both numerical calculation and experimental measurement. In this study, the contact temperature calculation follows the method of Papyrin et al. [5], where after obtaining the contact temperature, T_c, a solution of the relative strength, $a\%$, with different impact velocities, v_p, can be calculated with Eq. (1).

The calculation results of the interaction of Al–Si powder with the mild steel substrate is shown in Fig. 5, where the fraction of bonded atoms, for a given particle, starts to increase at a certain impact velocity; proving that a critical velocity is needed to obtain a reliable bond between the impact particle and substrate. At the same impact velocity, the fraction of bonded atoms for a bigger particle is higher than that of a smaller particle. This is because bigger particles obtain a higher contact temperature at the same impact velocity. The "✖" marks in Fig. 5 indicate the minimum required velocities for interfacial melting, and are connected by a dotted line with an arrow. It is certain that the relative strength of particle/substrate bond has achieved a maximum value before interfacial melting occurs, which confirms that interfacial melting should not be included as a dominant bonding mechanism of Al–Si feedstock kinetic spraying.

Because of the lack of a effective method to estimate the maximum adhesive strength $\sigma_{\max}$ of a given particle to a substrate, only the comparison of the relative strength $a\%$ to experimental bond strength is shown in Fig. 6, where 25 μm (mean size of experimental feedstock) is used for particle diameter in the calculation process. A critical velocity is needed for both the calculated relative strength and the measured bond strength, and these two curves rise with increasing impact velocity. A lower critical velocity for the calculated value is seen, and the growth of the calculated curve is ahead of the

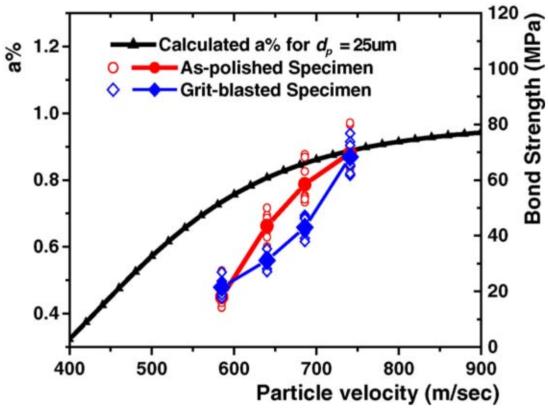


Fig. 6. Comparison between the experimental bond strengths to the calculated fraction of bonded atoms for the impaction of 25 μm Al–Si feedstock onto the mild steel substrate.

experimental one. Moreover, the calculations assume ideal conditions, where some phenomena, such as surface oxidation, micro-pore and other defects are ignored. Altogether, the calculated data is in good agreement with the experimental results, and thus this method can be considered an effective way of estimating the adhesive bond strength during high velocity impaction.

3.3. Surface roughness effect

In kinetic spraying, mechanisms of adhesion are still a mystery. Generally, mechanical anchorage, physical adhesion, and metallic interactions are involved in any kind of interfacial reaction. In nano-meter length-scale, the mechanical anchorage was explained as interfacial roll-ups and vortices by Grujicic et al.'s [6]. This interfacial reaction during high shear and high viscous flow along the interface was proved to be a dominant mechanism in the high speed particle/substrate bonding and enhancing. To determine the contribution of mechanical anchorage on interfacial bonding in micron/millimeter length-scale, the bond strengths on the two different roughness coating surfaces are measured in this study. It is believed that the rougher coating surface causes a higher mechanical anchorage; however, in our coating process it was easy to deposit the Al–Si particle onto the grit-blasted mild steel surface at low impact velocity, while it is failed to deposit onto as-polished surface at the same impact velocity. Unfortunately, only the first layer can be built up at such a low impact velocity since the over lapping deposition requires higher impact velocities [10]. The experimental results (Fig. 4) show that when the impact velocity is near the critical velocity, the bond strength of grit-blasted specimen is close to or exceeds that of the as-polished specimen. By further increasing the impact velocity, the bond strength of the grit-blasted specimen becomes less than that of the as-polished one. In this case, more fragments can be observed in the fracture surface of the as-polished specimen (Fig. 3). When the cohesive strengths of the two surface condition coatings are the same, the result shows that the adhesive strength of the as-polished specimen is closer to that of the cohesive strength; which is higher than the adhesive strength for the contact surfaces. In the high magnification micrographs, the micro-pores and defect can be found in the interface of the coating and rough substrate surface, while an intimate interface is observed in the as-polished specimen, as shown in Fig. 7. This can be seem as the reason for the lower bond strength in the grit-blasted specimens. It is observed that when the impact velocity is high (750 m/s), the bond strengths of the two kinds of coating surfaces is similar; which indicates that the micro-pores and defects on the interface are reduced by the high velocity impact. Therefore, in kinetic spraying the surface roughness has little effect on the interface bonding, and in most cases the mechanical anchorage

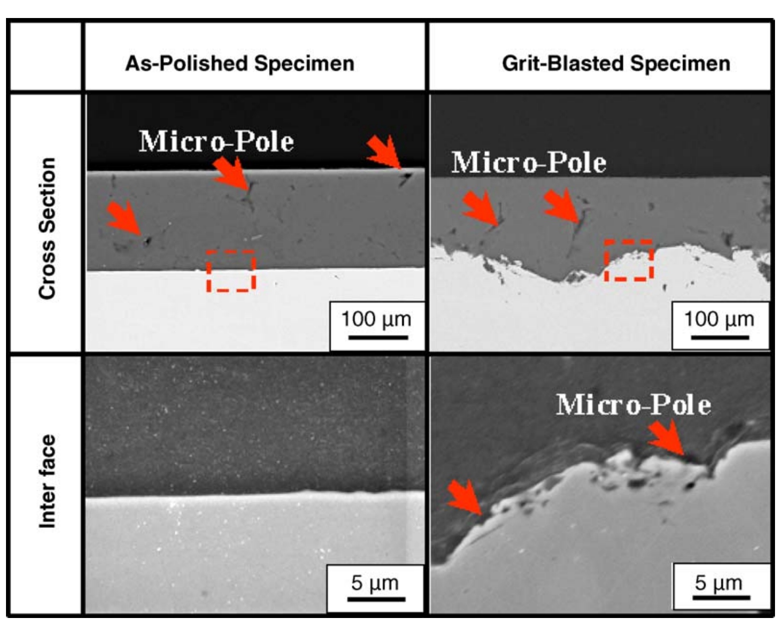


Fig. 7. SEM micrographs of the cross-section of Al–Si/mild steel coatings before the Stud Pull Coating Adherence Test.

in micron/millimeter length-scale is not a dominant mechanism for coating build-up.

4. Conclusions

In this study, Al–Si feedstock was deposited onto a mild steel substrate using different impact velocities. From the Stud Pull Coating Adherence Test, the bond strength of each coating was measured. A comparison between thermal spraying and kinetic spraying showed that the adhesive bond strength is much greater for kinetic spraying. Additionally, the cohesive bond strength was greater than the adhesive bond strength, thus causing the coating/substrate interface to fracture. It was determined that both adhesive and cohesive bond strengths are reduced by the micro-pores and defects produced during the spraying process. The bond strength increased with an increasing impact velocity. It can be argued that a higher fraction of bonds were generated between the two contact surfaces due to the higher impact velocity. Finally, the coating surface roughness did not significantly effect on the bond strength, and the mechanical anchorage in micron/millimeter length-scale is not a dominant mechanism for coating build-up in Al–Si/mild steel kinetic spraying.

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