

# Detection of oxygen at the interface and its effect on strain, stress, and temperature at the interface between cold sprayed aluminum and steel substrate



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## ABSTRACT

Aluminum powder particles were deposited on medium carbon steel substrate by cold spraying process. High resolution observation showed that the particles were severely deformed in solid state, whereas the substrate was hardly deformed. Furthermore, the particles were not bonded intimately to the substrate, and most of all, oxygen as well as thin gap were clearly detected along the interface of particle/substrate. Based on the observations, the impacting behavior of a particle on a substrate as well as the influence of the oxide film was modeled. The oxides covering the surface of metallic powder particles and the substrate significantly affect the impact and deformation behaviors of particle and substrate, and consequently the values of strain, stress, and temperature at the interface in the numerical simulation.

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## 1. Introduction

Aluminum and titanium have excellent corrosion resistance in many environments due to the stable, continuous, and protective surface oxide, which form spontaneously and instantly when fresh aluminum/titanium surface is exposed to air due to their high reactivity to oxygen [1–3]. However, they pick up easily oxygen in conventional thermal spray processes using heating up and melting feedstock materials. The thickness of the oxide depends on the two simultaneous parabolic processes, oxygen dissolution and oxide scale formation [4,5]. Accordingly, dynamic oxidation of sprayed particles in thermal spray processes can promote the oxidation and form amorphous metallic oxide [6].

The melting or semi-melting of feedstock materials in conventional thermal spray processes also causes undesirable phase transformation, high residual stress in coating layer, heating of substrate as well as oxidation [7–10]. Therefore, kinetic spray processes, such as cold spray [10–15], high velocity air fuel spray [16–18], kinetic metallization [19,20], and warm spray [3,21,22] using solid-state feedstock powder particles with high kinetic energy have been extensively investigated to minimize or

eliminate the detrimental effects. In the processes, metallic powder particles such as aluminum and titanium are accelerated by the supersonic gas flow or the high temperature gas flow. Since the processes were developed, however, the bonding mechanism of sprayed metallic powder particles onto substrate has been a critical issue [7]. Sprayed particles in the processes experience fast heating and cooling rates of about  $10^9$  K/s, high strains of 10 or even more depending on the location within the particles [10,12,21–24]. It is well known that bonding of sprayed particles onto substrate can be achieved when the velocity of sprayed particles ( $V_p$ ) exceeds a critical value, the so-called critical velocity for bonding ( $V_{cr}$ ). In other words, upon collision with substrate, the kinetic energy of sprayed particles should exceed the yield stress of sprayed particles [25]. Over the critical velocity, however, several different bonding phenomena have been suggested, and they can be divided mainly into three groups as the followings (even though some researches mentioned the possibility of no melting phenomenon because the occurrence of melting depends on the combination of sprayed particles and substrate [26–31]):

- (1) through surface adhesion by interface instability inducing physical anchoring effect [13,28,32].
- (2) through partial melting and fusion of materials in heavily deformed region [7,9,10,14,32–37].

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(3) through fracture of the surface oxide(s) covering sprayed particles and substrate [12,15,28,38–45].

Regardless of the mechanism, there is a common point that the compaction, deformation and plastic flow of sprayed particles under high pressure should remove the surface oxide(s). However, experimentally, a modest increase in oxygen content after cold spraying using helium gas which is generally used to increase the velocity of sprayed particles [11], and thin oxide layer on the atomic scale even at severely deformed and well-bonded region between sprayed particles and substrate were observed by high resolution analysis [6,46,47], which can affect the deformation behavior of sprayed particles in the process. Therefore, in this study, the detection of oxygen and its effect on strain, stress, and temperature at the interface as well as bonding formation between cold sprayed aluminum and steel substrate was extensively studied along the interface of a whole aluminum particle and the substrate.

## 2. Experimental

### 2.1. Cold spraying

Cold spraying was carried out using a home-made cold spray system (Jimei University, PR China). The specially-designed nozzle has a throat of 2.7 mm in diameter and expansion ratio of 8.8 with a divergent section length of 130 mm. Based on the capacity of the cold spray system, Air was used as a driving gas at a pressure of 3.0 MPa and a temperature of 500 °C because the relatively high temperature was used to promote the deposition of relatively large aluminum particles. However, taking into account the high scanning speed of spray gun (1000 mm/s) and the very short residence time of particles in hot gas, it is considered that there should be no in-flight oxidation, or it would be much limited. Argon was used as powder carrier gas at a pressure of 3.1 MPa. The standoff distance from the nozzle exit to the substrate was 30 mm. Commercially available pure aluminum powder particles with an average size of 52 μm and near-spherical shape were used as feedstock powder. A medium carbon steel (0.45 wt.% C) was selected as substrate, and mirror-polished prior to spraying. A single scan of the spraying gun was carried out on the substrate in order to make single-deposited particles.

### 2.2. Microstructural observation

As-sprayed particles were observed by high resolution SEM and then fabricated into TEM samples by a focused ion beam SEM device (FIB-SEM, FEI Quanta 3D) equipped with dual (ion and electron) beam and an electron dispersive X-ray spectroscopy (EDX) system. Two different FIB-SEM imaging modes, i.e. secondary electron (SE) and ion-induced SE (IS) by FIB, were used in this study. Thin lamellae were observed by a high resolution TEM (FEI Tecnai F20) with a scanning mode (STEM) and an EDX system, and a spherical aberration ( $C_s$ ) corrected TEM (JEOL JEM-2100FCS) with a STEM mode and an EDX. During high resolution TEM observation, every TEM sample was cooled with liquid nitrogen to avoid or minimize microstructural changes in the irradiated area.

### 2.3. Numerical simulation

The impacting behavior of a particle on a substrate was modeled using a commercial software (ABAQUS) with the Lagrangian formulation [48]. An axisymmetric model was used as shown in Fig. 1a. The Dynamic–Temperature–Displacement–Explicit procedure available in ABAQUS was used to clarify the effect of heat conduction. In the simulations, the width and height of the substrate were taken by 5 times larger than the particle diameter for

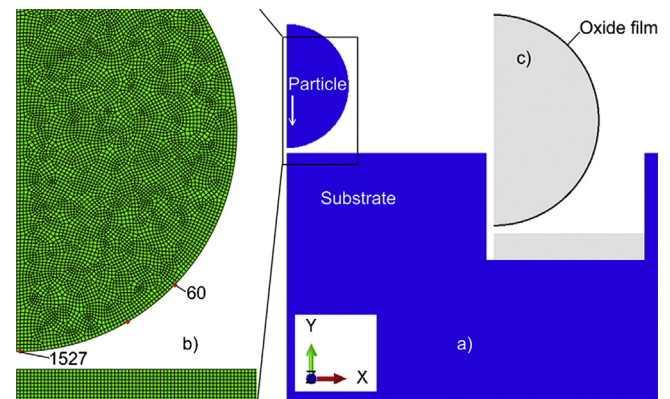


Fig. 1. Schematic diagram of the geometric model of particle impact (a) and meshing without oxide film (b) and with oxide film (c) in a resolution of mesh size being 1/100 particle diameter.

the sake of reducing the amount of elements and calculation time. The basic meshing size was 1/100 particle diameter as shown in Fig. 1b. As for the thickness of the oxide film, it is normally less than 100 nm, possibly 150 nm thick as Kang et al. [48] reported for the Al powder (mean size 65 μm) with one week room-temperature exposure to air. We have tried to change the oxide film thickness as 300 nm, 200 nm and 100 nm, which caused the considerable effect as observed in the previous study. However, the capability of the used computer is limited when using 100 nm thick. Hence, as a first approximation, 0.2 μm (one mesh size) was taken as the thickness of  $Al_2O_3$  as shown in Fig. 1c to investigate the influence of the oxide film. For the particle and substrate material models, the material deformation was described by the Johnson and Cook plasticity model, and the Johnson–Holmquist plasticity damage model (JH-2) was used for oxide film ( $Al_2O_3$ ) [49].

As marked on particle surface in Fig. 1b, node 1527 represents the center-bottom region and node 60 represents the well-bonded or largely deformed region. The changes of the nodal temperature (TEMP), equivalent plastic strain (PEEQ), equivalent stress (Mises) and shear stress (S12) with impacting time were investigated to enlighten the experimental findings.

## 3. Results and discussion

### 3.1. Solid state impact and deposition of aluminum powder particles

Fig. 2 shows SEM images of particles deposited on steel substrate. In cold spraying, as the temperature of sprayed metallic particles is below the melting point, aluminum can be deposited in solid state. When the sprayed particles are deposited on steel substrate, they are strongly impacted on steel substrate and severely deformed, while the substrate deforms little. As a result, the jetting-out phenomenon happens near the rim region of the particle. This solid state impact and bonding of aluminum powder particles in cold spraying is different from those happens in the melted state, such as the warm spraying using a low flow rate of nitrogen and the high velocity oxy-fuel processes [49]. The particles melted during conventional thermal spraying are splashed on substrate, or forms disk-shaped morphology. Therefore, compared the cross-sections of deposited particles, as the melted particles are much flatter than those deposited in solid state, one can easily distinguish aluminum particles deposited in solid state from those in liquid state. Fig. 2a therefore shows clearly, as already well known, that the sprayed aluminum particles in cold spraying are deposited in solid state, which is important to know the bonding mechanism in cold spraying and is discussed later.

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