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New insights into the coating/substrate interfacial bonding mechanism in cold spray

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ABSTRACT

A new theory was proposed to explain the interfacial bonding mechanism of hard Ni coating onto soft Al substrate. The experimental results indicate that the metal-to-metal contact and the consequent metallurgical bonding at the coating/substrate interface were absent in the single particle depositing but could be achieved in the full coating deposition. Based on this, it is proposed that the particle peening effect breaks the cracked oxides that remained at the coating/substrate interface into nano-pieces and promotes further deformation of materials. Thus, the pores caused by the bridge-like oxides at the interface are filled and the discontinuous metal-to-metal contact is achieved.

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Cold spray as an emerging coating technique has been developing for decades since its discovery in the 1980s [1]. In this process, powders are accelerated by the supersonic driving gas through a convergent-divergent nozzle and impact onto the substrate at a very high velocity to form the coating. Metals [2], metal matrix composites [3] and even pure ceramics [4] are able to deposit onto similar or dissimilar substrates with cold spray [5–7]. Unlike in conventional thermal spray, the feedstock used in cold spray does not cross the melting point during the coating formation, hence the inevitable defects appearing in the thermal sprayed coatings, e.g. oxidation, thermal residual stress and phase transformation, can be avoided in the cold sprayed coatings [8].

Bonding mechanism in the cold sprayed coating and at the coating/substrate interface is always a focal topic [8–16]. Mechanical interlocking and metallurgical bonding are believed to be the main bonding mechanisms in cold spray. Mechanical bonding is represented by non-chemical reaction, in which hard particles are mechanically trapped by the soft substrate material to form the interlock [12,13, 17–23]. Metallurgical bonding is the consequence of chemical reaction occurring at the inter-particles and coating/substrate interfaces, which requires the oxide-free interface and metal-to-metal contact. It normally provides high bonding strength and has been experimentally observed through the evidence of intermetallic or amorphous phases formed at the interfacial region or dimple-like dilute feature appearing at the fracture surface [9,13,24–28].

Currently, the most acceptable theory to achieve the oxide-free interface is that the outward metal jet as a result of the viscous plastic flow extrudes the cracked oxides from the interface to form the metal-to-metal contact [8,16]. This hypothesis can be used to explain the oxide removal in certain particle/substrate combinations, e.g. Cu onto Cu [29] or Al onto Al [30,31], where particles and substrate experience significant thermal softening at the interfacial region. However, in the case of hard particle depositing onto soft substrate, e.g. Ni onto Al, particle deformed slightly without metal jet formation [17]. Therefore, in this case, it is reasonable to consider that there must be other reasons for inducing the metal-to-metal contact and the consequent metallurgical bonding. In this study, well-designed experiments of cold sprayed Ni onto Al were conducted to clarify the bonding mechanism at the coating/substrate interface. Based on the experimental findings, a new theory to explain the partly oxide-free coating/substrate interface was proposed, which is applicable to the cold sprayed hard coating onto soft substrate.

The powder used in this study is spherical Ni powder (ECKAGRANULES, Batch: 9704) with the averaged diameter of 32 μm measured by the laser diffraction particle size analyser (Mastersizer 2000, MALVERN Instruments, UK). Al substrates with polished surface were employed for single particle and full coating depositions. Cold sprayed coatings were produced by a CGT K3000 system (LERMPS, UTBM, France), which consists of a computer control system, a powder feeder, a compressed high-pressure gas system, a gas heater, and a de-Laval nozzle (MOC, type 24 of CGT). The nozzle has a convergent and divergent length of 52.4 and 120 mm, respectively. The nozzle throat and outlet diameters are 2.7 and 7.8 mm, respectively. The pressure and

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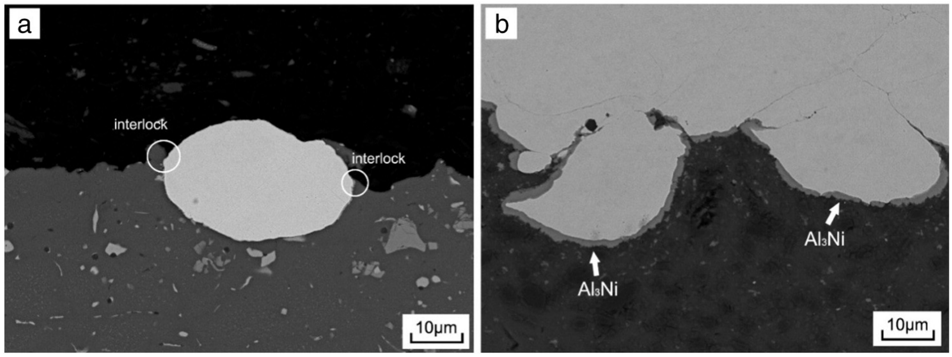

Fig. 1. Cross-sectional images of a single Ni particle (a) and coating (b) depositing on Al substrate after anneal treatment.

temperature of the propelling gas at the nozzle inlet were set as 2.8 MPa and 600 °C, respectively. The standoff distance between the nozzle exit and the substrate surface was set as 30 mm. The gun traverse speeds to produce the single particle deposition and full coating were set as 500 and 100 mm/s, respectively.

The interfacial bonding features were studied by evaluating the Ni—Al intermetallic phase formation after sample annealing and also by analysing the fracture surface morphology after tensile test. The samples for anneal treatment were heated to 400 °C, and held at this temperature for 15 min and then furnace cooled down to room temperature. The coating/substrate fracture surfaces were obtained by a tensile tester (IC ESCOFFIER, Estotest 50, France). The samples were adhered on the tensile test rods by the commercially available adhesive glue (FM1000 Adhesive, Couche Sales, LLC, USA) with a maximum tensile strength of 59 ± 3 MPa. Then, the assembled samples were pulled off with a cross-head speed of 1.56 mm/min. For the single particle sample, when failure took place at the interface between the substrate and glue layer, some deposited particles would be detached from the substrate and left on the glue layer. For the full coating sample, tensile test had to be done for several times until the fracture happened from the coating/substrate interface because failure firstly occurred from the coating inside. Scanning electron microscope (SEM) (JEOL, JSM-5800LV, Japan) equipped with an EDX unit (Oxford Instruments INCA system, UK) was used to characterize the single particle and coating microstructures.

Fig. 1 shows the cross-sectional images of a single Ni particle and coating depositing onto Al substrate after anneal treatment. In Fig. 1a, it is clearly observed that particle flattened after impacting and deeply penetrated into the substrate with some interlocking features forming at the rim. At the particle/substrate interface, no intermetallic phase was formed, which suggests the absence of intimate metal-to-metal contact between the particle and substrate. In the case of full coating deposition shown in Fig. 1b, however, the coating tightly bonded with the substrate, and an Al_3Ni intermetallic layer confirmed by EDS line-scan was formed at the coating/substrate interface. It is known that the formation of intermetallic phase is a consequence of chemical reaction across the interface, which requires the intimate metal-to-metal contact between two metals [17,32]. Therefore, the results shown in Fig. 1 indicate that the metal-to-metal contact can be achieved in the full coating deposition but absent in the single particle deposition.

Furthermore, Fig. 2 shows the fracture contact surface of a Ni particle and coating after they were detached from Al substrate. In the case of single particle deposition shown in Fig. 2a, it is seen that the contact surface was smooth and clean without any sign of metallurgical bonding. However, for the full coating deposition shown in Fig. 2b and c, Al material from the substrate can be clearly observed on the fracture surface, which means that the coating/substrate bonding strength at these zones was higher than the ultimate tensile strength of Al. Considering this finding, it is plausible to suggest that the discontinuous metal-to-metal and high-strength metallurgical bonding occurred at the coating/substrate interface.

Currently, the most acceptable hypothesis regarding the metallurgical bonding mechanism is that the outward metal jet as a result of adiabatic shear instability extrudes the cracked oxide film which originally exists on the particle and substrate surfaces from the interface and results in the metal-to-metal contact and thus metallurgical bonding. In our work, in the single particle deposition, Ni particle deformed slightly without any formation of metal jet. Therefore, according to aforementioned hypothesis, there will be no motive force to clean the cracked oxides and induce the metal-to-metal contact. This fact was proved by the absence of intermetallic phase and the smooth fracture surface at the interface shown in Figs. 1a and 2a. However, it is interesting to find from Fig. 1b and 2b that the metal-to-metal contact and the consequent metallurgical bonding took place at the coating/substrate interface when the full coating was deposited although without metal-jet. Therefore, it is sensible to consider that the aforementioned hypothesis is not applicable in this case and there probably exists another phenomenon triggering the metal-to-metal contact.

A new theory to explain the metal-to-metal contact and metallurgical bonding occurring in the full coating deposition was proposed. Fig. 3 shows a schematic of the coating/substrate interface evolution during the coating deposition process. As can be seen from Fig. 3a, in the single particle deposition, the oxide films originally existing on the particle and substrate surfaces are broken and cracked due to the high-velocity impact. These cracked oxides form a bridge-like structure and result in

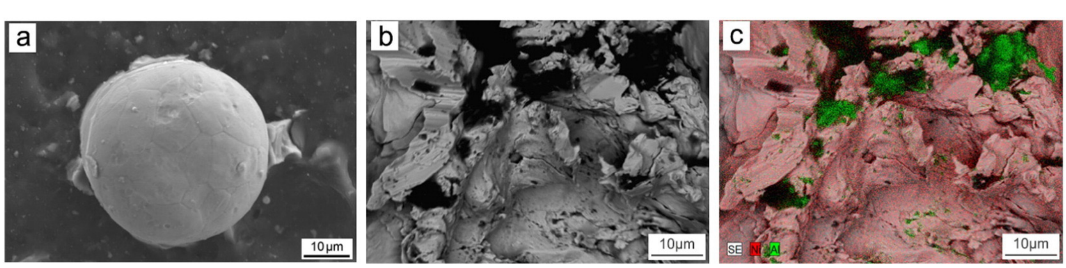

Fig. 2. Fracture contact surface morphology of a Ni particle (a), coating (b) and the corresponding EDS mapping (c) after they were detached from Al substrate.

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