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Full Length Article

## Improving interfacial, mechanical and tribological properties of alumina coatings on Al alloy by plasma arc heat-treatment of substrate



Guoliang Hou<sup>a</sup>, Yulong An<sup>a,\*</sup>, Xiaoqin Zhao<sup>a</sup>, Huidi Zhou<sup>a</sup>, Jianmin Chen<sup>a,\*</sup>, Shuangjian Li<sup>a,b</sup>, Xia Liu<sup>a,b</sup>, Wen Deng<sup>a,b</sup>

- <sup>a</sup> State Key Laboratory of Solid Lubrication, Lanzhou Institute of Chemical Physics, Chinese Academy of Sciences, Lanzhou 730000, China
- <sup>b</sup> University of Chinese Academy of Sciences, Beijing 100049, China

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#### ABSTRACT

Plasma sprayed ceramic coatings can be used to improve the mechanical properties and wear resistance of aluminum alloys, but there are still some challenges to effectively increase their interfacial adhesion. Thus we conducted plasma arc-heat treatment (PA-HT) of Al alloy substrate before plasma spraying, hoping to tune the microstructure of Al<sub>2</sub>O<sub>3</sub> coatings and improve their interfacial strength as well as mechanical and tribological properties. The influences of PA-HT on the microstructure of alumina coatings were analyzed by X-ray diffraction, transmission electron microscopy and scanning electron microscopy, while its effect on mechanical and tribological properties were evaluated by a nano-indentation tester and a friction and wear tester. Results demonstrate that a few columnar  $\delta$ -Al<sub>2</sub>O<sub>3</sub> generated on substrate surface after PA-HT at 200–250 °C can induce the epitaxial growth of  $\gamma$ -Al<sub>2</sub>O<sub>3</sub> grains in Al<sub>2</sub>O<sub>3</sub> coatings, thereby enhancing their interfacial bonding. Besides, elevating substrate temperature can help alumina droplets to melt into the interior of substrate and eliminate holes at the interface, finally increasing the interfacial anchorage force. More importantly, no interfacial holes can allow the heat of droplets to be rapidly transmitted to substrate, which is beneficial to yield smaller crystals in coatings and greatly enhance their strength, hardness and wear resistance.

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#### 1. Introduction

7075 aluminum alloy is widely used in aerospace and automobile industries, due to its high strength to weight ratio and ease of fabrication [1–3]. However, this Al alloy usually exhibits poor wear resistance, oxidation resistance and corrosion resistance, which is harmful to its reliability and lifetime [4,5]. Therefore, it is imperative to apply a variety of surface engineering technologies to modify the surface microstructure and properties of the Al alloy, thereby broadening its application scope. Among various surface engineering technologies, plasma spraying is of special significance, due to its low cost and high flexibility; and actually plasma spraying technique is applicable to fabricating Al<sub>2</sub>O<sub>3</sub>-based coatings with good mechanical properties and oxidation resistance as well as excellent wear resistance and corrosion resistance [6–8]. To date, it still remains a major challenge to minimize the differences in

the thermophysical and chemical properties of ceramic coatings

In terms of the deposition process, plasma sprayed coatings are built up by successive impacts of molten powders on the substrates, and the resultant individual splat can be considered as a basic cell [19–21]. This implies that the interfacial bonding strength of the plasma sprayed ceramic coatings should be highly

and metal substrates in order to improve the interfacial bonding strength between the two types of heterogeneous materials [9,10]. Plasma sprayed Al<sub>2</sub>O<sub>3</sub>-based coatings on steel substrates exhibit a low adhesion strength of about 15–30 MPa [11–13], and the same coatings deposited on Al or Mg alloy substrates with higher thermal expansion and thermal conductivity even exhibit an adhesion strength of less than 10 MPa [14,15]. The direct consequence of the poor adhesion strength is that most coatings undergo peeling-off and failure initially at the coating/substrate interface [16]. To overcome this bottleneck, some researchers resorted to sand blasting of metal substrate, introduction of bond coatings, and optimization of spraying parameters, but they harvested few in enhancing the interfacial bonding strength of ceramic coatings therewith [15,17,18].

<sup>\*</sup> Corresponding authors.

E-mail addresses: csuayl@sohu.com (Y. An), chenjm@licp.cas.cn (J. Chen).

dependent on the flattening behavior of the initial impinging droplets as well as the topography and composition of the substrate surface [22,23]. Reminded by this perspective, some researchers attempted to conduct preheating of the substrates in order to manipulate the substrate surface composition and the splat shape, thereby improving the adhesion strength of plasma sprayed ceramic coatings. Chandra et al. [24] and Valette et al. [25] found that the preheating of a low carbon steel substrate at elevated temperatures results in the transition of the splat shape from a distorted one to a disk-like one, which is favorable for increasing the interfacial contacting area and for promoting the formation of oxide transition layers with similar crystal structure on the substrate surface. As a result, the Al<sub>2</sub>O<sub>3</sub> coating plasma sprayed on the low carbon steel substrate preheated at 350 °C exhibits a bonding strength of up to 60 MPa.

However, it remains a challenge whether the preheating technique is applicable to Al alloy substrate, which exhibits a lower melting point, a poorer anti-oxidation ability and a higher thermal conductivity than steel substrates [26]. Besides, Chraska and King [27] found that as the steel substrate is properly preheated, many nano-columnar crystals can be formed in the initial impinging yttria-stabilized zirconia (YSZ) splats obtained by plasma spraying traditional powders. Unfortunately, there has been little work reported about whether these refined crystals can exist in the whole ceramic coatings, not to mention their influence on the mechanical and tribological properties of coatings. Namely, it is still one of the greatest dreams of the thermal spraying community to acquire Al<sub>2</sub>O<sub>3</sub> coatings with a high bonding strength on Al alloy substrate, especially with better mechanical properties and wear-resistance.

In the present research, we adopt a plasma arc to preheat 7075 Al alloy substrate at different temperatures and then apply plasma spraying to deposit alumina coatings from traditional feedstock. The flattening behavior of the alumina droplets and the influence of plasma arc-heat treatment (PA-HT) on the morphology and composition of the Al alloy substrate are reported. It mainly deals with the coating-substrate bonding mechanisms as well as the microstructure, mechanical properties and wear resistance of the ceramic coatings deposited on the Al alloy substrates with and without PA-HT. This research, hopefully, is to provide a feasible method for preparing ceramic coatings with high interfacial bonding and mechanical properties on Al alloy substrate.

#### 2. Experimental procedure

#### 2.1. Plasma arc-heat treatment

Prior to PA-HT, the 7075 Al alloy substrate with a dimension of  $\emptyset$ 24 mm  $\times$  7.7 mm.  $\emptyset$ 25 mm  $\times$  40 mm 100 mm × 25 mm × 8.5 mm was sandblast by silica to increase the surface toughness ( $Ra = 2.13 \pm 0.02 \,\mu\text{m}$ ), or mechanically ground with emery papers and polished with diamond paste to a mirror surface ( $Ra = 26.2 \pm 4.9 \,\mathrm{nm}$ ). The as-roughened or aspolished Al substrates were then heated with a plasma arc (power: 36 kW) to  $100 \pm 10^{\circ}$ C,  $200 \pm 10^{\circ}$ C,  $250 \pm 10^{\circ}$ C and  $300 \pm 10^{\circ}$ C, respectively, which were detected on the lateral faces of substrates by an infrared temperature measurement gauge. In order to quickly and precisely heat the substrate to a pre-set temperature, the moving speed  $(20 \,\mathrm{mm/s})$ , distance to the plasma arc  $(120 \,\mathrm{mm})$ , and time of exposure (5-30 s) to the plasma arc must be properly adjusted. More importantly, besides removal of the water physically adsorbed on substrate surface, some oxide layers with similar crystal structure to alumina could be formed on the substrate surfaces during the PA-HT process under the abovementioned conditions.

**Table 1** Plasma spraying parameters.

Items	Values
Primary gas (Ar) flow rate (L/min)	40
Secondary gas (H <sub>2</sub> ) flow rate (L/min)	0.2
Current (A)	600
Voltage (V)	60
Powder gas flow rate (L/min)	12
Powder feed rate (rpm)	20
Gun speed (mm/s)	1000
Spray distance (mm)	100
Injector angle (degrees)	90

#### 2.2. Preparation of alumina splats and coatings

An APS-2000 plasma spraying system (Institute of Aeronautical Manufacturing Technology; Beijing, China) operated by an IRB 2400/16 robot (ABB, Sweden) was performed to deposit traditional alumina powders (size:  $15-45\,\mu m$ , fused  $2O_3$  splats and coatings. Ar and H2 gas were used as the primary and secondary plasma gases, respectively, and the optimum spraying parameters are listed in Table 1.

# 2.3. Measurements of bonding strength, shear strength and mechanical properties of coatings

The bonding strength and shear strength between the coatings and Al alloy substrates (Ø25 mm × 40 mm or  $100 \, \text{mm} \times 25 \, \text{mm} \times 8.5 \, \text{mm}$ , roughened or polished) was measured with a CMT 5205 universal testing machine (SANS testing Machine Co., Ltd.; Shenzhen, China) according to the ASTM C 633-01 standard [28] and HB 5494-91 standard [29], respectively, with which a commercial E-7 epoxy resin glue was used as the binder. Moreover, the coated area on the working surface ( $100 \, \text{mm} \times 25 \, \text{mm}$ ) of Al alloy substrate (100 mm  $\times$  25 mm  $\times$  8.5 mm) is 7 mm  $\times$  25 mm. The adhesion strength ( $P_L$ , in N/mm<sup>2</sup>) of the as-sprayed coatings is calculated as:  $P_L = 4F_I/\pi D^2$ , where  $F_L$  (in N) is the maximum load and D (in mm) is the diameter of the contact zone between the coating and substrate. The shear strength ( $P_S$ , in N/mm<sup>2</sup>) of the as-sprayed coatings is calculated as:  $P_S = F_S/2A$ , where  $F_S$  (in N) is the maximum load and A (in  $mm^2$ ) is the single-side overlap area (7 mm × 25 mm). A NHT 02-05987 nano-indentation tester (CSM, Switzerland) was performed to measure the mechanical properties of the as-deposited coatings. The nano-indentation measurements were conducted on the polished surfaces of each coating at a maximum load of 40 mN, a loading rate of 80 mN/min, an unloading rate of 80 mN/min, and a pause time of 10 s.

### 2.4. Friction and wear test

Friction and wear tests were conducted with a ball-on-disk tribometer (CSM, Switzerland) at room temperature (about  $20\,^{\circ}$ C). The Al alloy substrates (Ø24 mm × 7.7 mm) with the polished alumina coatings were used as the lower specimens, and commercially available  $Si_3N_4$  balls (Ø6 mm) were used as the upper specimens. The sliding tests were conducted at an amplitude of 2.5 mm, a normal load of 10 N, a constant sliding speed of 10 cm/s, and a sliding distance of 120 m. The friction coefficients were recorded by the attached computer. Upon completion of the friction and wear tests, the wear volume losses of the alumina coatings were automatically measured with a three-dimensional (3D) non-contact surface mapping profiler (ADE Corporation; Massachusetts, USA). The wear rates are calculated as W = V/SF, where W is the wear rate in mm<sup>3</sup>/Nm, V is the wear volume loss in mm<sup>3</sup>, S is the total sliding distance in m, and F is the applied load in N.

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