Laser-induced surface texturing of metal or organic substrates for structural adhesive bonding

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Abstract. Laser-matter interaction is commonly described regarding three main factors: laser beam, materials and environment. Conversion of absorbed energy via collision process into heat is the most important effect that occurs during laser interaction. Short-pulsed laser beam induces fast transition from the overheated liquid to a mixture of vapor and drops which allows the ablation of micrometric layers. Specific patterns can then be achieved using scanning and automation technology also called laser texturing.
New materials with specific properties such as endurance life and/or lower environmental impact attract emerging technologies such as thermal spraying. However, adhesive bond strengths have to be high enough to play a key role in surface properties. A clean surface to enhance mechanical interlocking is a key element. Mechanical and physico-chemical bond strength for thick coatings elaborated by thermal spray process can then be developed using laser.
The aim of the present paper is to show the potential of such emerging treatments through new results using various thermal spray processes (thermal spraying as well as cold spraying). Metal or organic materials were investigated implementing various powders.

Introduction

Adhesion strength of thermal spray coatings strongly depends on substrate preparation, preheating temperature, morphology and composition of the substrate surface [1[2]. A clean substrate surface prior to coating is the baseline for promoting substrate-coating interface which depends on desorption of the pollutants adsorbed on the surfaces and the droplet wetting [3]. Surface contaminations such as oxides, carbon or oils have to be removed from a metallic surface before its final use as they change the physico-chemical behaviors and/or surface topography [4]. Among conventional techniques, degreasing and grit-blasting are used in most cases. Degreasing agents involve to chemical modifications of the surface while grit-blasting modifies the surface morphology by creating a uniform roughness thus promoting a mechanical anchorage of the incoming particles to the substrate [5]. This technique is very effective except for ductile materials where potential microcrack nucleation could be observed [6]. In addition, grit inclusions can occur decreasing the adherence of the subsequent coating. Then new technologies such as laser tools are developed to adjust the coating/substrate adhesion. Recently, laser tools have been shown to improve surface behaviors of materials as surface treatment techniques (for cleaning purposes, topography modification, heating treatment, etc.) [7[8[9[10[11]. The benefits are as follows: easy automation, localized treated area, three dimensional treatments and great flexibility. A controlled ablation technique may involve modifications in topology for all types of materials like glasses, ceramics, polymers and metals [12]. A specific laser tool fitted to the material to be treated (in terms of wavelength, pulse duration, spot size and pulse energy) added to a scanner for 3D shape modification can promote mechanical adherence for thick coating elaborated by thermal spraying. Those parameters affect the topography but also the material microstructure due to the heat flux which can be absorbed during the treatment according to the pulse duration and the laser-matter interaction

[**13**][14]. For this, short pulse duration (10^{-10} – 10^{-15}s) is needed to localize the laser interaction on the extreme surface.
Adhesion is related to the nature and strength of the bonding forces between two materials such as ionic, covalent, metallic, hydrogen and Van der Waals forces [15]. The mechanical interlocking appears to play a dominant role in thermal spraying [16]. Considering the substrate as well as the powder composition, specific spray processes can then be selected according to the temperature and the kinetic of the jet. Particles impact the substrate with various behaviors in terms of temperature (the particles can be molten or un-molten) and speed (from 200 to 1200 m/s). Specific relationships can then be observed at the interface substrate/particles inducing various adherence. By spraying molten particles, matter fulfills the substrate morphology before solidifying whereas cold spraying involves un-molten particles thus interface occurs by plastic deformation of the materials (particles and substrate). Different adherence behaviors can then be measured particularly according to the substrate surface morphology.
To illustrate the influences of process and substrate topography on coating adherence, different materials couples have been sprayed and analyzed. In order to compare the process used, metallic (Al, Cu) and ceramic powders (ZrO_2-Y_2O_3) were sprayed onto metallic substrates (stainless steel, aluminum alloy) or organic composites implementing Atmospheric Plasma Spraying, cold spraying and wire arc spraying. Conventional method (grit-blasting) and laser surface texturing have been compared through adhesion bond strength tests.

Experimental Procedure

Materials. Various material couples have been treated to explore all the benefits of such surface pre-treatment. In order to investigate several industrial applications (automotive, aeronautic, aircraft component reinforcement, etc.), diverse materials and processes have been tested as presented in Table 1. Whatever the considered system, the substrates were 25 mm diameter and 10 mm thick buttons.

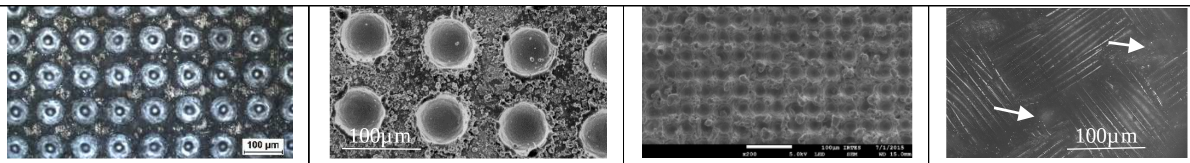
Table 1: Material couples tested

Substrates	AISI 304L	AS7G0.6	AW5083	C-Epoxy (TSC) C-PEEK (TPC)
Powders	ZrO_2-7Y_2O_3-1,7HfO_2 (Praxair - ZRO 236-1 ; d_{50} : 63µm)	Al5056 (d_{50} : 30µm)	Cu-Al_2O_3 (50-50 w. %) (Dycomet - K-01-01 ; d_{50} : 28.4µm)	Cu-05T wires (Tafa)

Surface treatments. According to the material couple, thermal and cold spray processes have been selected for elaborating coatings. Zirconia coatings (YSZ) were produced by atmospheric plasma spraying (APS), using a F4 torch (Sulzer-Metco). The torch was mounted on a XYZ robot (ABB robot). Samples were rotated in front of the torch while the torch operated a vertical movement allowing a homogeneous 300µm thick coating. Samples were cooled down to room temperature by an air cross-jet perpendicular to the substrate. Coatings were deposited using standard thermal spray parameters as presented previously [17]. For ductile metallic materials (aluminum alloys), 300µm thick coatings were elaborated by cold gas spraying. Aluminum coatings were obtained via N_2 dynamic spray process using a Kinetics 3000 gun (CGT GmbH, Germany) whereas copper deposits were sprayed using air low pressure cold gas spray process (Dymet 423, Dycomet, Netherland). In both cases, the spray gun is a converging-diverging nozzle mounted on a XYZ robot (ABB - Switzerland). The gas conditions were selected to obtain several in flight particles velocities: 600m/s (using N2) or 350m/s (using air). Standard gas parameters as well as spray conditions are selected [18]. Considering the organic substrates, coatings were elaborated using a wire-arc spray process

(TAFA 9000, Praxair, USA) implementing pure copper wires and standard spray parameters [19]. Whatever the technology used, all experiments have been carried out at 90° spraying angle.

To promote coating adhesion, several processes have been performed. Grit-Blasting (GB) was used to obtain homogeneous substrate roughness with varying mean values according to the treated material. As innovative process, laser surface texturing was performed using a pulsed fiber laser device (Laseo, Ylia M20, Quantel, France) with wavelength 1.064 µm, pulse duration 100 ns, maximum power 20W and frequencies varying from 20 up to 100 kHz [20]. The laser beam is circular with 60 µm diameter at the focal point and a Gaussian energy distribution. The surface patterning technique consisted of series of equidistant lines covered with a number of holes as a spotted surface. For this, the scanner stops the laser beam and pulses are generated to build holes. Then, several shapes can be defined according to the number of lines fixed in X and Y directions and the number of pulses (Fig. 1). It modifies the surface contact area of the substrate. Particularly, the optimal cavity dimensions must be adapted depending on the spray process, the powder and the melt particle viscosity. In this study, holes were chosen as a simple pattern. The textured substrate could have numerous adhesion areas depending on the shape, height, orientation, distribution and density of holes. The study aims to have the hole volume equal to sprayed powder average volume and the hole diameter larger than particles mean diameter. Then, holes of various diameters (50-60µm) and depths (20-80µm) were designed and the distance between holes (along X and Y) was studied (from 80 to 300µm).



AW5083 substrate	304L substrate	AS7G0.6 substrate	TPC composite

Fig. 1: Example top Views for Al alloys, AISI and composite surface patterning

Characterization methods. The adhesion of thermal-sprayed coatings is highly related to the substrate surface area, surface morphology has to be precisely measured. Conventionally, standard roughness parameters (Ra, Rz, etc.) characterize grit blasted surfaces. As many adhesion areas can be obtained on textured surfaces, depending of the shape, height and density of holes, a more complex morphology can be observed including pattern geometry and "additional" surface roughness (spatters and recast material). In order to compare surface pretreatments on coating adhesion, a new characterization approach has been investigated. The characterization of the morphology of the pre-treated samples without coating was performed by optical microscopy (Moz2 Zeiss) and by Scanning Electron Microscopy (SEM Jeol JSM-6400). Both surface and cross section observations were performed using cross section images, the interface area is determined by image analysis using the fractal approach following 3 steps : (1) a small area is considered from a previous microscopy image, (2) threshold treatments define the substrate/coating boundary and (3) ImageJ software computes the developed interface edge length [11]. For grit-blasting, the adhesion area is directly related to the interface edge length. Similarly, for laser surface patterning, it is possible to compute the hole interface length from a cross-section view and to calculate the interface area for one pattern (Dundurs equation). The pattern enables to evaluate the complete adhesion area by taking into account both textured and non-textured areas (Fig.2). An interface or adhesion area ratio R can then be defined which represents the degree of the interface area compared with an equivalent planar (as Ra for profilometer analysis) [20]. Regarding the surface treatment, several wavelength patterns can then be obtained (in-between 1.7 and 7 for laser texturing, compared with 1-2 for grit-blasting). Ten image analyses for each treated surface (different samples) were carried out to estimate R to statistically provide reliable data.

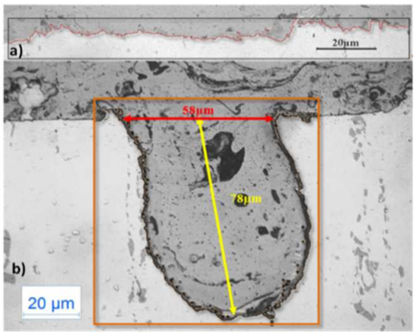


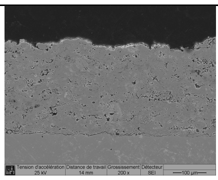
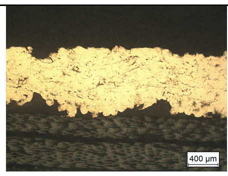
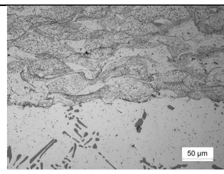
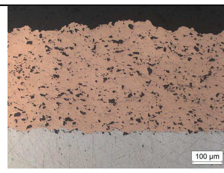
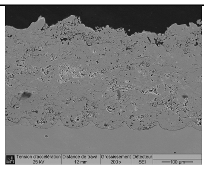
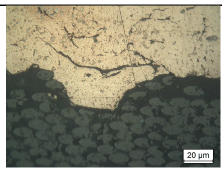
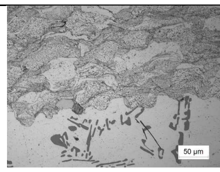
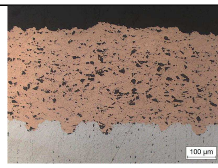
Fig. 2: Image Analysis Adhesion Area: a) grit-blasting surface and b) laser-textured surface

The coating adhesion was performed in a similar manner as that described by the DIN EN 582 (ASTM Standard C633). 25 mm diameter button samples were joined with cylindrical counter-parts using an adhesive agent (FM1000) [18,19]. A constant displacement (1.026 mm/min) was applied to the counted parts with a tensile test machine (50kN – 500mm ESCOTEST) up to complete failure. The maximum force was then attributed to the adherence at the interface calculating the ratio between the force and the tested area [20]. At least, 5 measures were performed for each condition test to calculate a mean adherence value.

Results and discussion

Whatever material behaviors as well as spraying process, different morphologies at the interface substrate-coating can be observed (Table 2).

Table 2: Coating cross-sections

	YSZ /304L - APS	CU/COMPOSITE – WIRE ARC	Al/Al - CS	Cu-Al_2O_3/Al - LPCS
Sand blasted substrate				
Textured substrate				

Conventionally, grit-blasting induces rough and irregular interfaces whereas texturing produces a regular structural surface. Nevertheless, whatever the sprayed material (using different spraying process), interface morphology is perfectly homogeneous. A regular contact seems ensured between both materials (substrate and coating) particularly for textured material where the holes appear completely fulfilled. By optimized texturing (in terms of hole morphologies: depth, diameter), interface behavior can then be promoted.

To evaluate the effect of surface preparation, tensile adhesion tests were performed on previous coating-substrate samples. Figure 3 represents the adherence values for all material combinations function of surface ratio factors (R).

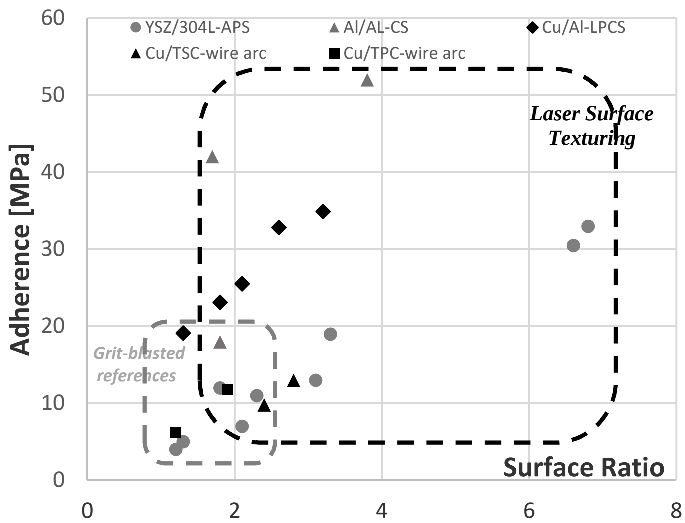
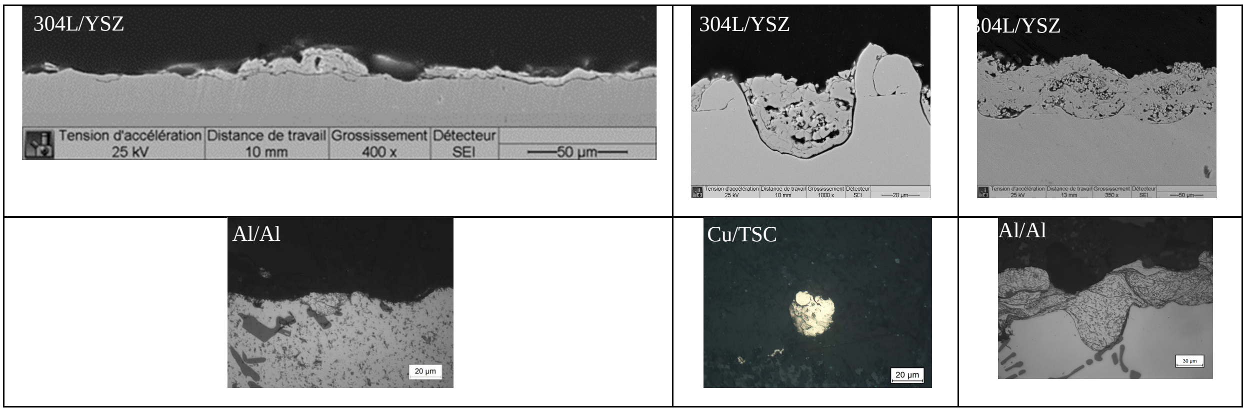


Fig. 3: Coating adherence related to surface ratio considering several material couples sprayed by various processes (grit-blasted references noted by circle).

At first, an increase in the coatings adherence appears with the surface ratio systematically. Second, laser texturing develops surface ratio (surface ratio from ~2 to 7) compared to the grit blasted surfaces (limited to a surface ratio around 1-2). Consequently, laser texturing can improve coating adherence simply by increasing the interface length. Larger contact between coating and substrate increases mechanical bonding at the interface. Moreover, holes can lock coating part when traction adhesive tests occurred. As observed in Figure 4, cohesive and adhesive failures can be distinguished according to the substrate surface morphology.

For sand-blasted surfaces, particles detached completely from the interface following the interface morphology (Fig. 4-a). Inversely considering textured surfaces, coatings can be trapped in keyholes. For large matrice (200 or 300µm inducing low surface ratio ~2-3), hole might be considered isolated. So plane spacing where the cracks can propagate easily is identified. Then, it is easier to go above holes going through the coating to let particle locked in holes (Fig. 4-b). A mixed mode failure can then appear combining adhesive (coating-substrate interface) and cohesive (through the coating) adhesion. By contrast for dense pattern (100µm, high surface ratio), cracks are stopped at each hole and have difficulties to go around. Some particles are trapped in the hole and others next to hole are linked to the substrate (Fig. 4-c). The coating cohesion is in this case lower than the interface adhesion.



a b c

Fig. 4: Micrographies of interfaces after Tensile Test for Grit-Blasted Surface (a) and textured surfaces: adhesive (b) and cohesive failures (c)

Conclusions

The application of a fiber laser to create micro-patterning on metallic surface in order to promote coating adhesion has been studied. The findings are summarized as follows:

1. The influence of substrate surface topographies has been studied and the effects on the coating adhesion have been determined. It has been shown that the Wavelength pattern has an effect on the adherence due to mechanical anchoring which stops crack propagation.
2. Textured surface with optimized hole morphology allows to obtain an interesting adhesion value, higher than that generally observed with conventional pretreatments.
3. Holes create obstacles for crack propagation. Cracks go through the coating (following the plane direction) increasing debonding energy necessary.
4. Laser texturing can be applied on different materials and benefits can be measured whatever the process carried out.

Other benefits of laser technologies are their low-processing times, high flexibility, precision and low-environmental impact, which could encourage industrial coating manufacturers to implement them in thermal spray processes.

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