

# Ceramic/fluoropolymer composite coatings by thermal spraying—a modification of surface properties

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## Abstract

Even if polymer coatings are widely used for industrial applications, their performances are often limited by a poor scratch resistance or a high water and gas permeability. To overcome such limitations, the composite coating of ceramics with polymer can be a solution to be adopted. Thermal spraying may help to achieve this objective, thanks to its ability permitting to deposit many kinds of materials with an important range of coating thicknesses. The aim of this paper is to develop composite coatings (fluoropolymer/ceramic) by plasma spraying. Due to different thermal characteristics of the initial materials (PTFE or PFA polymers and  $\text{Al}_2\text{O}_3\text{--TiO}_2$  ceramic), three types of powders injections are elaborated. To compare these three conditions, microstructure, surface wettability and wear resistance deposits behaviour were characterized.

$\text{Al}_2\text{O}_3\text{--TiO}_2$ /fluoropolymer (PTFE or PFA) composite coatings are characterized by a well-melted ceramic matrix in which rounded polymer particles are randomly distributed. The as-sprayed polymer particles kept at the coatings surface can however be spread thanks to a thermal treatment at 350 °C during 15 min. Unlike the PFA, a large PTFE loss was noticed during the spraying process. Thus, the highest polymer/ceramic ratio is observed in the coatings made with the separated injection of the  $\text{Al}_2\text{O}_3\text{--TiO}_2$  and PFA powders. The lowest friction coefficient is also measured for this coating.

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## 1. Introduction

Thermal spraying is a well established means of forming relatively thick (50  $\mu\text{m}$ ) coatings. A wide variety of materials can be thermal sprayed for applications ranging from gas turbine technology to electronics industry. Thermal spray consists in melting material feedstock (wire or powder) to accelerate and propel heated particles towards a substrate where rapid solidification and thickness build-up occur. Thus, a heat source and a means of accelerating the material are required. The high temperature for melting is achieved either chemically (through combustion) or electrically (by arc) which plays the role in accelerating the particles. Sprayed on the target substrate, the deposit is also built-up by successive impingements of individual flattened particles or splats.

Thermal spray processes were classified according to the type of heat source. Based on combustion, flame spraying and high velocity oxy-fuel spraying (HVOF) are quoted and characterised by a low temperature (below 3000 °C) achieved within the combustion flame and a velocity varying between 50 (flame) and 2000  $\text{m s}^{-1}$  (HVOF) according to the technology. In electric arc spraying, several techniques can be employed as two wire electric arc, atmospheric plasma (APS) or vacuum plasma spray (VPS) and selected according to specific elements such as temperature, velocity, environment or material throughput. Depending on their principle, many jet characteristics can also be obtained and a wide diversity of materials are used as thermal spray feedstock like metals, intermetallics, cermets, ceramics, glasses as well as polymers. Due to their thermal characteristics (thermal conductivity, melting temperature, evaporating temperature) as well as particles morphologies (size, crystallisation, etc.), specific techniques will be also employed to spray materials. For example, ceramic coatings are more often elaborated by plasma technology, whereas

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those of metallic materials are sprayed by HVOF or wire-arc and polymer by flame.

In recent years for example, polymers coatings have found an ever growing use in several industrial applications such as corrosion protection, chemical or wear resistance [1,2]. High performance thermoplastics and many others are widely used (poly-ether-ether-ketone (PEEK), polyphenylene-sulphide (PPS), polyester (PET, PBT), polyamide (PA), polyethylene (PE), polyurethane (PU), fluorocopolymers) [3]. However, the low characteristic temperature and the irregular morphology of the particles with a large grain size induce some difficulties for thermal spraying. Whatever the technique used to spray polymers, a more drastic control of the molten particles is required. Two main parameters can be distinguished between thermal conductivity and particle size. The thermal characteristics being very low for all the organic materials, an important thermal gradient inside the particle is developed which must be as short as possible between the surface and the middle to permit a good splashing on the substrate surface. The second one concerns the particle sizes due to the small temperature variation between the molten and the degradation state of such elements. Taking into account their low thermal conductivity, shorter the particle size is, more complete the melting is. The microstructures of thermally sprayed deposits are based on the solidification of many individual molten droplets. Then, the splat morphology (which occurs when a droplet of molten material strikes the surface and solidifies) is directly linked to the molten state of the material. Of course, many other parameters influence the mechanical or physical aspects of splat formation depending on the spreading of the molten droplets and the interaction with the substrate. These characteristics are affected by the splat viscosity, the particle velocity, the surface tension, the substrate surface profile as well as the temperature and the diameter of particles. Then, if thermal spraying technology can be controlled for metallic or ceramic material, spraying polymers presents more difficulties considering all these aspects. But, if organic materials present interesting properties especially for surface applications (chemical resistance, low friction), yet such characteristics can involve some drawbacks for thermal spraying. In the case of PTFE for example (which presents important advantages concerning the low friction component), the non-adherence and its high viscosity at the molten state induce some problems for splat formation. This way, the thermal spray of polymers needs adaptations (preheating of the substrate, powder injection, etc.) and particularly when mixing with another material with very different properties.

Moreover, the properties of the polymer coatings are often limited by poor scratch resistance and high water and gas permeability [4]. Even if no tests have already been realized by thermal spraying, some composite blocks using fillers (nanoparticles of  $\text{SiO}_2$ ,  $\text{ZnO}$ ,  $\text{Al}_2\text{O}_3$ ) can be elaborated by compression to overcome both limitations [5,6]. For example, to improve this property, many kinds of PTFE matrix composites have been made in combining PTFE and

different nanoparticles as PPS,  $\text{ZnO}$  or  $\text{Al}_2\text{O}_3$  [7–10]. This way, mechanical resistance can be then provided by filler materials whereas the surface properties can be supported by the organic materials. However, even if all desired surface properties are obtained, most applications concern only the material surface (wear resistance, abrasion resistance, etc.). So, it is necessary to develop a composite coating which corresponds to the aim of this paper.

The work of this paper describes thermal spraying (specifically plasma spraying) of fluoropolymers (PTFE and PFA) and ceramic ( $\text{Al}_2\text{O}_3$ – $\text{TiO}_2$ ) powders using three types of injection to form polymer–ceramic composite coatings on aluminum substrates. Microstructure and wear resistance behaviour of the different coatings obtained with optimised parameters were studied and compared. The wettability of coatings was also analysed.

## 2. Thermal sprayed ceramic–polymer composite coatings

Polymer coatings are often obtained by flame spraying [11,12]. However, when ceramic–polymer composite coatings are considered, plasma spraying is preferred because of its higher energetic jet [13]. To ensure melting of ceramic particles without degrading the polymer, three types of the injection of composite powders into plasma jet can be studied: blended-powder injection, cladded-powder injection and separated injection of both types of materials.

In the case of composite coatings produced by a mixture of polymer and ceramic powders, the hydrogen percentage in the  $\text{Ar-H}_2$  plasma jet is an important spraying parameter. The average temperature of particles increases with the increase of hydrogen content [14]. This influence is due to the effect of this gas on the convective heat transfer coefficient of the plasma jet, which enhances the heat flux to the particle surface. However, owing to the limitations of internal heat conduction within the polymer [15], this heat flux cannot be conducted fast enough to the interior of the particle. The temperature at the particle surface rises much more rapidly than in the core and causes rapid decomposition. For example, there is no volume loss of a 60- $\mu\text{m}$  PMMA particle when it is immersed in pure argon, while in an  $\text{Ar} + 5\% \text{H}_2$  plasma, the volume loss is 35% only after 50  $\mu\text{s}$  of entering the plasma jet [16]. This phenomenon shows that the hydrogen rate must be optimized to reach a balance between maximizing the melting of ceramic particles and minimizing the decomposition of the polymer. An adjustment of the spraying distance is also necessary to achieve a satisfactory microstructure (as regards porosity, polymer distribution in the coating) and deposition efficiency [17]. A too weak spraying distance decreases the time of plasma–particle interaction, which does not enable a well heat transfer from plasma to the particles surface. If the spraying distance is too high, particles are cooled down and resolidified during their flight into plasma jet [18]. This parameter controls the impact temperature of particles [19].

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