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Columnar suspension plasma sprayed coating microstructural control for thermal barrier coating application

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ABSTRACT

Suspension plasma spraying (SPS) is used to perform enhanced YSZ coating with columnar microstructure for thermal barrier coating (TBC) applications. By combination of plasma flow, substrate preparation, suspension formulation and injection or coating kinematic management it is possible to tune SPS coating structure from widely-separated columns to a significantly more compact columnar structure. Among these parameters, substrate roughness control, combined with an adapted coating growth velocity, are identified as the most relevant. An analytical approach is presented to describe columns growth based on coating image analysis. It allows to give the expression of the lateral and normal growth speeds responsible of the columnar structure.

1. Introduction

Thermal barrier coatings (TBCs) are widely used in aeronautic industry in order to prevent thermal degradation and oxidation of the component of hot section of gas turbines [1]. Particularly, turbine blades, located after the combustion chamber, are submitted to high operating temperatures over 1100 °C. TBCs are complex multilayer stacks composed of an insulating ceramic top coat, mostly made of 7–8 wt.% yttria partially stabilized zirconia (YSZ), coated on an alumina forming metallic bond coat for oxidation protection of the nickel-based superalloy substrate. Main issue for gas turbine is to increase operating temperature which necessitates more efficient TBCs [2].

Nowadays, YSZ ceramics top coats are mainly performed using Electron Beam Physical Vapor Deposition (EB-PVD) [3]. Columnar microstructures, resulting from germination of condensed material on a metallic substrate, offer high thermal compliance during operation [4]. It induces high lifetime for these TBCs mainly attributed to the stress accommodation capability of the columnar structure. Stress results from thermal coefficient expansion (CTE) mismatch between YSZ and metallic components. However, due to columnar voids, acting as heat conduction paths, and due to the dense microstructure of columns, a thermal conductivity, higher than 1.5 $W\,m^{-1}\,K^{-1}$, is measured in that case [2]. This is a clear limitation identified for next generation of gas turbines. Moreover, EB-PVD process presents high cost compared to more industrially generalized technics such as thermal spraying.

Thermal spraying and most specifically atmospheric plasma spraying (APS) is a widely used coating process to design highly porous YSZ TBCs with rather low overall cost. In APS process, YSZ feedstock particles are injected into the plasma jet by a carrier gas and are then spread over the metallic substrate as splats [5]. Particles are flattened and adopt a more or less disk shape depending on substrate temperature and roughness [6,7]. A lamellar microstructure is obtained by stacking splats during process. As they experienced various thermokinetical treatments inside the plasma jet, particles present molten, semi-molten or un-molten states, additionally to important cooling rate (>10^7 K/s) when they are flattened onto the substrate. As a consequence, plasma spray coatings develop a 3D inter-connected micro crack network which leads to a porous coating favorable for a low thermal conductivity [5,8]. Nevertheless, lamellar TBCs performed by APS present lower thermal lifetime properties compared to EB-PVD mainly due to the extension of microstructural defects parallel to the bond coat [9]. In order to overcome this limitation and to increase lifetime, dense vertically cracked (DVC) microstructures can also be

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performed by APS for which cracks are developed perpendicularly to the substrate all along the coating thickness [10]. DVC microstructures have demonstrated better thermal lifetime than lamellar ones. It is assumed that cracks enhance strain tolerance to thermal mismatch [11,12]. Nevertheless, DVC structures generally present lower porosities and exhibit vertical cracks acting, as well as columns in EB-PVD, as heat conduction path probably leading to lower insulating coatings [13].

Even if by using APS process it is possible to vary to some extend coating morphologies (homogeneous, DVC), microstructure still remains lamellar. Changing APS lamellar microstructure to a finely structured sub-micron scaled porous coating is challenging without decreasing the size of starting particles. But, injection of particles with diameter lower than 5 μm requires high carrier gas flow rate which significantly perturb the plasma jet resulting in low deposition efficiency and a poor coating quality [5]. Nevertheless, to prevent this perturbation nanosized powders can be used in an agglomerated form, but in that case the range of accessible microstructure is quite reduced, often leading to a bimodal structure made of un-molten nanosized particles imbedded in a fused matrix of molten material [14,15].

The use of suspensions as carrier media for nanosized particles resulting in Suspension Plasma Spraying (SPS) allow to overcome the injection limitation of nanosized particle into the plasma jet, leading to nanostructured coatings [16]. Momentum for injection into the plasma flow is ensured by liquid dropplets which become greater than that of plasma momentum allowing a good penetration of particles in the core of the plasma. Ethanol-based and water-based are used for suspension formulation. Consumption of enthalpy from plasma flow can be decreased using volatile solvent as ethanol [17]. Particles are then heated and accelerated, alone or as agglomerates, before substrate impingement in molten or semi-molten state. Lower standoff distances are required for SPS compared to APS to prevent in-flight re-solidification of molten nanoparticles [18,19]. SPS has demonstrated its ability to perform coatings with multi-scaled porosity [20] as well as different morphologies: homogeneous, vertically cracked and columnar [18,21,22]. Last one is promising for thermal compliance as well as for thermal insulation [23,24]. Even if mechanisms of formation of SPS columnar microstructure are not yet completely understood, some studies proposed preliminary explanations. Particularly, Van Every et al. [22] explained that columnar growth features during SPS are induced by the deviation of the smallest particles for which trajectories follow the plasma flow modified at the substrate vicinity. Close to the substrate, plasma flow direction is significantly modified becoming almost parallel to its surface [25]. According to numerical computations, particles in range 1 to 5 μm seems to be affected by the direction of plasma flow [26]. Assuming that the smallest particles follow the plasma jet trajectory close to the substrate, they impinge its surface with a strong parallel velocity component. At the end, coating results in both lateral and normal growth around asperities.

As thermomechanical properties and thermal insulation of TBCs are strongly affected by their microstructures, the aim of this study is to give some rules to describe and manage morphology of SPS columnar structure. Modification of direct parameters such as substrate roughness or plasma condition is explored. Moreover, the effect of coating growth speeds by varying suspension load and relative torch/substrate velocity (named here linear speed) on columnar structure is investigated.

2. Materials and methods

2.1. Design of experiments

In order to vary columnar structure morphology and to identify relevant parameters, different spraying conditions are chosen. As previously described, columnar structure is mainly driven by the deviation of the smallest particles into the plasma jet close to the substrate leading to combined normal and lateral growth of the coating. Therefore, two plasma conditions, summarized in Table 1, are first studied, differing from enthalpy and velocity values. Secondly, in order to illustrate the effect of the amount of suspension (both material and solvent) injected into the plasma, two different feed rates 25 g/min and 44 g/min are chosen. Then, the substrate roughness, the load of the suspension and the torch linear speed are changed. All the coatings performed are described in Table 2 where labels A and B are related to the plasma gas mixture, subscripts 25 and 44 described the feed rate used and subscripts Ra, L and V described changes in substrate roughness, load of suspension and torch linear speed respectively. Deposition efficiency, also presented in Table 2, corresponds to the ratio between the mass of resulting YSZ coating and the mass of YSZ injected into the plasma jet. Errors on deposition efficiencies are taken as standard deviations between different samples and can be calculated using Eq. (1) where x represents deposition efficiency and where n is the different measurements.

$$\Delta x = \left[\frac{\Sigma (x - \bar{x})^2}{(n-1)} \right]^{1/2} \quad (1)$$

2.2. Coating production

SPS coatings are performed at atmospheric pressure using F4-VB plasma gun with a 6 mm internal diameter nozzle provided by Oerlikon-Metco using plasma gas mixtures A and B (see Table 1). Plasma characteristics are calculated at the exit of the plasma torch using ALEX software from Limoges University based on thermodynamic calculations. Spraying distance is fixed at 50 mm in order to provide a sufficient particle dwell time into the plasma jet without significant inflight re-solidification of molten particles. Linear speed of plasma torch is adjusted from 1000 mm/s (A_{25}) to 1500 mm/s ($A_{25\text{-}V}$). Cooling is ensured by a combination of both compressed air and cryogenic liquid CO_2 jet. 7 wt.% YSZ ethanol-based commercial suspensions with mean particle diameter (d_{50}) lower than 300 nm from Treibacher Industry AG™, are used to performed SPS coatings. The initial load of 25 wt.% can be decreased by ethanol dilution. Suspensions are stored into a pressurized vessel, stirred using a magnetic agitator and are injected radially. Coatings are performed on stainless steel and aluminized Hastelloy®X substrates. Roughness of different substrates can be adjusted using sandblasting from 0.6 μm to 1.5 μm.

2.3. Microstructural analysis

Coating microstructural analyses are performed on polished cross sections (stainless steel and aluminized Hastelloy®X substrates) and top surfaces (stainless steel substrates only) using Scanning Electron Microscopy (SEM). A Leo 435VPi SEM is used with secondary electron detector for SEM surface analysis and back scattered electron detector for cross sections with 3 magnifications (500×, 1000×, 10 000×). Statistical analysis on columns size is performed on SEM top surface using ImageJ, a public domain

Table 1
Plasma gas mixtures characteristics at the exit of the torch on the jet axis.

	A	B
Mixture	$Ar/He/H_2$	Ar/He
Mass gas rate (g/s)	1.5	1.5
Power (kW)	32.2	22.4
Enthalpy (J/kg)	2.2×10^7	1.5×10^7
Temperature (K)	12 100	11 400
Velocity (m/s)	2906	2332